

Referee#1

We would like to thank the Referee for very detailed reading of the manuscript and numerous suggestions. In the process of revision we tried to follow his recommendations.

“It is a notable contribution to extending existing fluorescence and water vapor Raman lidar techniques and an interesting topic for the lidar community. The context and study goal are clear, and the results don’t show major errors and omissions. However, there are many presentation styles, grammar, and typo errors. Is this a haste-writing? Therefore, I recommend it to be considered for publication after some revisions.”

In the revised manuscript numerous corrections of style were introduced.

“In general, the structure of this manuscript needs to be improved. Some part of the abstract is read like a conclusion, such as line 31-38, while the conclusion is too detailed about the results discussion without extracting the main points. The introduction also can be more converging to the aims of this study, and a better context is needed between paragraphs, e.g. line 71-72, it seems water vapor just come out without connection with the above.”

In revised manuscript we strongly decreased Abstract and Conclusion following Referee recommendations.

“For the scientific part, the typical values of fluorescence depolarization ratio (line 277-280), the hygroscopic growth cases in BL(line 307), and the assumption of “change of aerosol composition” (line 315) as well as “change in particle morphology may affect the depolarization ratio of fluorescence” “at RH about 90%, δF increases up to 70%.” (means more spherical higher δF ?), these above probably should be discussed, because it seems the results or shreds of evidence do not support them well.”

We agree with Referee, that the additional supported evidences of observed effects are desirable. However, it is not always possible. For example, observed variations of fluorescence capacity should be related to variation of the smoke composition. However, to support of this statement, the independent in situ smoke sampling inside the smoke layer is needed, which was not available at the time. Some effects are observed in this study for the first time, and we are not able to provide the complete scientific explanation. For example, increase of δF with RH inside the PBL. It occurs, definitely, not due to the spherical shape, but, can be due to increase of molecules mobility inside the particle. Unfortunately, we have no information about rotational recombination time or about fluorescence life time of molecules in atmospheric aerosol, to provide quantitative analysis. Suggested mechanism is just one of possible, and additional studies are needed. Still, we think that it is important to present the results to community, it may stimulate others for research in this field.

Details:

“The title “**Derivation of aerosol fluorescence and water vapor Raman depolarization ratios...**” is easy to mis-understand, “**Derivation of depolarization ratios of aerosol fluorescence and water vapor Raman backscatters...**” might be better.”

We agree, the title is modified

Abstract:

“Line 16: “The total scattered power of” means to “the total backscattered power of?””

Yes, corrected

“Line 19: regarding the same source of smoke, where is this measurement taken place relative to Moscow, so it is at Lille?”

Yes, at Lille. We had two systems, at Moscow and at Lille

“Line 27: “...fluorescence at 513 nm while, in the upper...” to “fluorescence at 513 nm, while in the upper”

Corrected

“Line 36: should be “the strongest...””

Corrected

Line 38-40: suggest to re-write this sentence since it is difficult to understand.

Corrected. Now it is “As a result, depolarization ratio at the water vapor Raman is sensitive to the presence of strongly depolarized fluorescence backscattering and can be used for evaluation of corresponding uncertainty of the water vapor mixing ratio (WVMR) measurements.”

Introduction:

“Line 57: “In fluorescence spectroscopy..” a comma is missing. And what is the motivation to introduce the anisotropy?”

Corrected. We modified the sentence, now it is “In the fluorescence spectroscopy, the polarization state of emission is described by the anisotropy (Lakowicz, 2006), introduced as:”

“Line 65-67: check the grammar.”

Checked

“Line 67: “minimal” should be “maximal”?”

No, this is minimal. Observed depolarization should be higher than 33%.

“Line 68: it is unclear. Why the molecule rotation could increase the depolarization?”

If rotation is faster than the fluorescence life-time, the emission is completely depolarized. This is widely used technique for viscosity measurements, so we just provide reference to Lakowicz book.

“Line 80: Could the authors please be more specific about this issue “fluorescence still remains the issue”?”

Changed for “fluorescence still remains the source of uncertainties, especially when the water vapor mixing ratio (WVMR) is measured inside the smoke layers in the upper troposphere”

“Line 84: “weakly” to “weak””

Hm..., looks like weakly is correct...

“Line 149: “For both channels” a comma missing”

Corrected

Results :

“Line 293: “High depolarization ratios” means particle depolarization ratios? at 532 nm?”

Yes, we introduced it in the beginning of section 2.

“Line 295: “particle depolarization” to “particle depolarization ratio”?”

Yes, corrected

“Line 355: The lowest”

Corrected

“Line 358: please give a definition of the “ratio””

Done

“Line 360: please explain how is the standard deviation obtained.”

For calculation of standard deviation we used the expression

$$s = \sqrt{\frac{1}{N-1} \sum_{i=1}^N (x_i - \bar{x})^2}$$

Where N is number of measurements x_i and \bar{x} is the mean value. This is standard definition and we are not sure, that it should be presented it in the manuscript.

“Line 406: the depolarization ratio of water vapor is due to the mixing with fluorescence?”

Yes

“Line 410-411: please check the grammar and re-write the sentence.”

We corrected the sentence

“Line 251, 312, 354, 355, 375, and so on...: “5 – 21 May” and “9-16 June” Please unify the format everywhere, and also the symbols for indicating the parameters. “1.8%÷2.0%” to “1.8%-2.0%”? “On 26-27 and 28-29 May the uncertainty” There are a lot of such format or typo issues, I will not point out one by one.”

In revised manuscript we use everywhere the AMT format “5-21 May”, “5-10%”...

Conclusion :

“Line 426-427 : I suggest to remove this sentence as there is no any loss without it. In general, make it more compact, and emphasize on the main findings and contributions.”

We agree. It is removed. Conclusion is strongly shortened.

Referee #2

First of all we would like to thank the Referee for the large amount of work he has done revising out manuscript. In the revised version, we tried to follow his recommendations.

“The English text will be improved once the proposed corrections are accepted and performed.”

The corrections suggested by Referee are introduced in the revised manuscript.

“Some citations are missing in various places and some others are proposed to be added (eg. lines 47, 81, 132, 224, 289).”

Citations are added

“It is not convincing that the detected aerosol layers correspond to smoke only, and not to other aerosol sources, as no air mass backtrajectory analysis with height has been shown. The authors should add these graphs in the supplement section.”

The smoke layers were identified by the fluorescence capacity. For the episodes considered it exceeded $2.5 \cdot 10^{-4}$, and no other aerosol type can provide it. The back trajectories for all considered cases show transport of smoke from North America (we added corresponding comment to the revised manuscript), but we would not like to overload the manuscript with additional figures.

“How convincing are the “*dry smoke events*” mentioned in the manuscript, when a complete air mass trajectory analysis is missing?”

In revised manuscript we removed “dry smoke layers” and use “layers with low water vapor content”. Water vapor is measured by lidar. Besides, high values of the fluorescence capacity are possible only in the absence of hygroscopic growth.

Specific comments

“*line 51*: The paper of Wang et al., 2023, has to be omitted as it refers to a multi-wavelength elastic-Raman-fluorescence lidar system and not to a single-channel lidar as mentioned in the manuscript, as this placement there is misleading.”

Done

“*line 119*: I would ask the authors to provide in a Supplement section a new figure detailing the transmission spectra (zoom in on the Transmission curves for T between 0-20%) of the filters at the 5 wavelengths mentioned, so that the readers can see (in detail) the overlapping transmission curves between these filters. Based on Veselovskii et al., (2023, Fig. 1) we can clearly see these overlapping regions, so the authors have to discuss on any induced errors in the detected fluorescence signals and their role on the accuracy of the retrieved aerosol parameters. “

Transmission bands of the interference filters are completely separated. Some overlap in Fig.1 (Veselovskii et al., 2023) corresponds to the transmission bands of the interference filter (IF) and the dichroic mirror (DM) used for separation: reflection of the DM starts to decrease near the long-wavelength edge of the IF. This effect is the strongest when 560 and 610 nm channels are separated, however it decreases the power at the 560 nm channel for less than 2%. This is

beyond accuracy of our calibration and so was ignored. In revised manuscript we added a phrase to the system description. Thus, probably no need to provide the Supplement section.

“line 216: See comments there (about the definition of the nv parameter etc.).”

Changed

“page 21: For clarity reasons limit the longitude up to 80° only (Fig. 2).”

Done