

First of all, we would like thank the Reviewer for careful reading our manuscript and suggestions. Below are responses to his comments.

Summary: The manuscript reports on fluorescence measurements of atmospheric aerosols with a multi-wavelength Raman lidar, where the interference filter in the water vapor Raman channel was replaced by a broadband filter around 466 nm. Although the study contains some interesting approaches, e.g. the possible synergy of combined measurements with multi-wavelength Raman lidar and fluorescence lidar, it is incomplete and too speculative at this stage and requires substantial extensions and improvements for a possible publication. For example, it is incomprehensible why the authors do not present aerosol events that could show the real strength of their modified lidar system (microphysical retrieval plus fluorescence), but only those that are actually not suitable. A little more patience would have been necessary here.

As mention by the Reviewer, combining the multiwavelength and fluorescence measurements can be a promising approach for aerosol characterization. But to implement it, some important questions should be answered first. These questions, in particular, are:

- Is fluorescence technique sensitive enough to be useful for lidar aerosol measurements, when part of the spectrum is selected by the interference filter?
- How the fluorescence signal is affected by the variation of the relative humidity and by the droplets formation?
- Is it possible to measure the fluorescence signal inside the cloud layer?

In our manuscript we tried to get answers for these questions and to demonstrate the feasibility of our system for fluorescence studies.

By today we have measurements performed during high aerosol loading, and combining of multiwavelength retrievals with fluorescence data is in progress. However this is a subject of separate study. We wouldn't want to add multiwavelength inversion to this one.

We should recall also, that when aerosol near the cloud base is considered, usually the aerosol extinction coefficients are quite low and traditional multiwavelength Raman technique does not work. In our study we consider cases with low aerosol loading and suggest approach, based on use of predefined aerosol models, for aerosol characterization.

Furthermore, the paper shows technical weaknesses in both the experiment and the analysis, and the interpretation of the measurements is highly speculative. For example, the fluorescence measurement has not been thoroughly calibrated,

Equation (9) for the fluorescence backscattering contains the ratio of efficiencies of fluorescence and Raman channels. The dichroic optics used, allows efficient separation of fluorescence and Raman signals, so main source of uncertainty is relative sensitivity of PMTs in the channels. To equalize sensitivities, the PMT from fluorescence channel was installed in the Raman one and by small adjusting of voltage the same value of nitrogen Raman signal was obtained. The cathode sensitivity of R9880U-01 PMT between 387 nm and 466 nm changes for less than 15%, thus we assume that sensitivities of PMTs in both channels are the same and only difference in transmission of interference filters was considered. We estimate that uncertainty of such calibration is less than factor 2, which is sufficient for our purpose, because relative variations of

fluorescence backscattering coefficient are considered. Corresponding comment is added to the text.

and no backward trajectories were used for aerosol typing.

In revised manuscript we discuss backward trajectories

Furthermore, the measurement results are discussed using relative humidity, although neither water vapor measurements with the lidar nor local radiosondes were available.

Yes, RH data were available only from radiosond in Belgium (95 km away). However in this study we don't analyze the hygroscopic growth. RH data are taken as qualitative only.

Interestingly, the authors themselves point out some of these weaknesses in their conclusions, they should fix them and then resubmit the manuscript.

We definitely understand all these weak points, still we think that this study presents new important results.

Major issues:

1. The calibration of the lidar was not performed with a spectral lamp (l. 169 ff), so the measurement trueness is questionable, and the authors are aware of this (l. 380 ff). Why was the calibration not performed? Nevertheless, the measurements are quantitatively evaluated and interpreted, this is not a consistent approach.

We have already responded this comment. Discussing the cross sections obtained, we emphasize, that these are only rough estimations.

2. The authors speculate about the presence of aerosol mixtures (l. 204 ff). This can only be investigated with spectrometric fluorescence lidars, if at all. But at least an analysis of the backward trajectories should have been performed.

In revised manuscript we added back trajectory analysis. Air masses pass Africa and particle depolarization ratio is high. So dust is predominant in aerosol mixture.

This also applies to the statements regarding the change of GF (l. 225 ff).

We are not able identify aerosol type for this case

3. Relative humidity is used for the interpretation of the measurements, although it is not known sufficiently for these purposes, especially for hygroscopic aerosol growth (l. 211 ff). Thus the interpretation is a speculation.

We don't analyze hygroscopic growth. We just say that RH is high at 1000 m and drops above 2000 m. The sonde data from England and from Belgium lead to similar conclusion.

4. Particle depolarization is not only a function of particle shape but also of particle size, this should be considered in the discussion.

Yes, depolarization definitely depends on particle size. But here we focus on the fluorescence. Analysis of dependence of depolarization on particle parameters is out of the scope of this study.

5. The whole microphysical interpretation (l. 244 ff) is pure speculation. Why did the authors not wait for aerosol measurement cases where they could have used the strengths of their multi-wavelength Raman lidar?

In situations, when aerosol extinction is low (for example when aerosol near the cloud base is analyzed) the multiwavelength Raman technique can not be used. So other approaches, allowing at least qualitative estimations of particle properties are needed. The estimations of particle properties, based on predefined aerosol models are widely used in remote sensing. In our study we used the aerosol models based on AERONET observations. Still we agree that such estimations need numerous assumptions, thus results obtained can be considered as qualitative only. Corresponding comments are added to revised manuscript.

Discussion paper

6. The reviewer is sceptical about the measurements in chapter 3.2, which are supposed to prove an internal mixture of aerosol particles and cloud droplets (l. 311 ff).

Reviewer 2 provided numerous comments, concerning “internal and external mixing”. So in revised manuscript we don’t use this terminology.

It is noticeable that the fluorescence signal associated with the cloud layers seems to be a function of the measurement height: below 1000 m very high ‘fluorescence’ values are found in clouds, around 1500 m slight increases, and above 1700 m elastic and fluorescence signals are uncorrelated. This may (but of course does not have to) indicate instrumental effects (height-dependent angle-of-incidence distribution of the backscattered photons). Are there measurement examples where liquid water clouds below 1000 m do not show increased fluorescence?

Yes, reviewer is right, height dependence of fluorescence is complicated and depends on aerosol loading. We think that this is result of water uptake by aerosol (aerosol dissolving, water shell forming...). We don’t see how instrumental effects can result in such profiles, because we had many aerosol observations without such “exotic” behavior at low altitudes (Fig.6 in this manuscript). Still in the presence of high RH elastic scattering and fluorescence don’t correlate, because water uptake by particles normally does not increase fluorescence significantly.

Minor issues:

1. The authors claim that lidars with spectrometers are less sensitive than those with standard detection channels (l. 61 ff). However, a comparison with published spectrometric measurements seems to contradict this. Please explain in more detail.

Transmission of the interference filters used is above 95%, while transmission of grating spectrometer with fiber input is definitely lower. This why we say that spectrometer is less sensitive. In revised manuscript we modified this phrase as

“However, sensitivity of such lidar spectrometers is lower when compared to the technique based on selection of fluorescence spectrum intervals with interference filters, because the transmission of modern filters exceeds 90%”.

2. The authors plan to reduce the bandwidth of the interference filter for fluorescence measurements by a factor of 2 or even 4 in the future (l. 404 ff). However, this would further increase the measurement duration, which is already very long. Please explain in more detail.

In Fig.2 we show fluorescence maps obtained with 2 min resolution at low aerosol loading. So we have resource to reduce the filter width. But our experience of fluorescence measurements (and data analysis) shows that we never have “too much” signal. So the phrase about bandwidth reduction is removed from revised manuscript.

Wording:

1. To speak of a ‘highly efficient lidar operation’ (l. 368) when in fact hour-long integration times are needed for fluorescence measurements is quite a stretch.

Changed for “efficient”

Type setting:

1. All variables in the running text and in the equations must be checked for correct math format. There are many formatting errors, for instance, variables are not italic (e.g., l. 133), or subscripts are italic (e.g., l. 119).

Changed

1. Figures 1 and 7 are of poor quality.

Why? We don’t think that these are of poor quality...

2. Figure 6, colors for beta₁₀₆₄ and beta_F are hardly distinguishable when printed.

We changed color of beta 1064 line for blue.