Response to Referee #1

First, we are very grateful to the Referee for accurate reading the manuscript and useful suggestions.

The paper is well written and deals with an important aerosol fluorescence lidar application. The authors show how spectrally resolved aerosol fluorescence information can be used to distinguish local boundary layer aerosol and smoke in the free troposphere (after long-range transport). I have only minor comments:

Page 1, line 27, smoke influence on cirrus clouds is nicely shown by Mamouri et al., ACP, 2023. Should be cited!

Done

Page 2, line 46 and 48: It is confusing that the fluorescence capacity depends on hygroscopic growth, and the fluorescence spectrum does not…. Please explicitly explain that this is related to the humidity dependence of the total backscatter coefficient.

Corresponding sentence is added.

Figure 2: larger letters are needed for the different panels (a), (b), (c)….. this holds for most figures.

Fonts are increased

The separation or division of the altitude range in PBL, free troposphere and UTLS is not satisfactory. One should better use words 'lower free troposphere' and maybe 'middle and upper troposphere'. The UTLS defines the region around the tropopause and frequently suggests interaction between the upper troposphere and lower stratosphere. However, you discuss and distinguish smoke in the lower to middle free troposphere and smoke in the middle to upper troposphere. So please state that accordingly. Avoid UTLS if you discuss smoke in the upper troposphere only.

We agree with reviewer. In the revised manuscript we use "Lower Free Troposphere (LFT) and "Middle and Upper Troposphere (MUT)".

Section 3.2.
Figure 5, …. (a) and (b) are too small, the symbols are too small, the legend is too small, everything is not easy to read (in the printout) and thus it is difficult to get the main message.

Figures are modified

By studying the figures, the question arises: Why is that? What are the reasons for the differences? Maybe the higher smoke layers are lofted by convective cloud activity, and the lower layers are just emitted and ascend as a result of sunlight absorption. Aging of smoke depends on the availability of gases (emitted together with the particles) and is faster when the humidity is high….. The probability for faster aging is higher in the lower free troposphere than in the typically dry upper free troposphere. With time and condensation of gases the BC fraction decreases typically towards a few percent, and at the same time the organic carbon fraction increases…. Can these arguments explain your observations better? At least as a reader I missed reasons for the findings

…dynamical aspects, injection aspects, transport aspects, chemical aging, cloud processing and aging…. Even if only hypotheses can be presented, this will stimulate further research.

These questions are indeed crucial, and our study offers valuable new insights. It is now well recognized that the chemical aging of smoke increases the organic carbon (OC) fraction, thereby enhancing fluorescence capacity. Our measurements reveal that fluorescence capacity—and thus the OC fraction—tends to increase with altitude. One possible explanation is that smoke aging in the upper troposphere may occur more rapidly, despite lower relative humidity. This aging process is influenced by various factors, including injection parameters and transport conditions. However, at this stage, we are unable to identify the exact mechanisms involved, and further research is needed to clarify these dynamics. Corresponding comment is added to the manuscript.

Figure 6 seems to support that different vertical transport phenomena are active for smoke below and above 8 km height above Moscow.

This is probably true not only for Moscow. Results obtained in Lille (manuscript is in preparation) demonstrate that smoke particles size increases with height. Thus, smoke particles in lower and upper troposphere have not only different fluorescence properties, but the microphysical parameters as well.

Figure 7: again nice results, clear differences, and the question arises? What controls these differences? Again, (a), (b), (c)…. too small. And different scales in (d) vs (e) and (f) is not helpful.

Figures are modified

Section 3.3 should be shortened. The article is quite long, and two extended case studies in Section 3.3 are probably too much for the reader.

In revised manuscript we shortened Section 3.3. Second case is removed.

I missed the depolarization ratio observations. Was the particle depolarization ratio always close to zero? It may have been significantly enhanced in the dry upper troposphere.

Unfortunately, at current configuration of the lidar, the depolarization ratio is not available.

The conclusion section does not mention the results in section 3.3, maybe one should remove section 3.3?

We shortened this section, but prefer to keep it, because it is important to demonstrate separation of smoke and urban particles based on their fluorescence spectra. Corresponding comment is added to Conclusion.

Figures 11 and 16: Because of the clouds it is difficult to see the pathways of the smoke transport, over the continents and over the ocean.

Yes, we agree. The figure is modified.

Page 16, line 332: smoke lidar ratios at 355 nm of down to 20 sr? Values of 35-60 sr for 355 nm.

In Ansmann et al (2021) variation of S355 is 35-50 sr, while Hu et al, (2022) report lower values (25-50 sr). Corresponding correction is introduced in manuscript.

A final remark: The probability for pure smoke is highest in the upper troposphere and therefore the fluorescence spectrum for this smoke may be used as reference spectrum.

From observations performed in Lille and Moscow we can conclude, that fluorescence properties of smoke in the lower and upper troposphere are different. As discussed above, these properties are influenced by the processes of smoke aging and transportation. So, by our opinion, we should use different smoke spectra for lower and upper troposphere. However, in our algorithm, the choice of the spectra does not influence significantly the results.