

Response to RC1

First of all, we would like to thank Referee for very detailed analysis of our manuscript. In the process of revision, we tried to follow his suggestions.

The authors use a fluorescence lidar to study the hygroscopic growth of urban aerosol at Lille, France. Interestingly, they report periods where the fluorescence backscatter coefficient is not affected by increasing RH and other periods where fluorescence quenching occurs, i.e., a decrease of the fluorescence backscatter with increasing RH. These are important findings because they imply that the fluorescence backscatter coefficient can not always be used to normalize the backscatter enhancement due to hygroscopic growth in not well mixed aerosol layers. Furthermore, the authors report extinction-to-volume and extinction-to-surface-area conversion factors based on the inversion of the lidar data. The applied methods are valid and well presented. The study is relevant and deserves publication in AMT after some minor revisions listed below.

Major comments:

As it remains unresolved which type of urban aerosol is affected by fluorescence quenching, it is desirable to gather as many information as possible on the episodes with and without fluorescence quenching. Probably, this question can not be resolved in this publication, but providing as many as possible information (e.g., a lidar ratio) on the aerosol situations observed during the 5 case studies will help future scientists to find patterns.

The values of lidar ratios and overlap heights for all episodes are added to the revised manuscript.

What about the occurrence of pollen during your observations? The fluorescence quenching cases are from May and June and might be linked to a certain pollen species?

The main pollen season in Lille occurs between March and April, though pollen episodes can also occur in May. Using fluorescence and depolarization measurements, we took special care to confirm that pollen did not contribute to aerosol backscattering during the studied episodes.

You provide backscatter hygroscopic growth parameters (γ_{β}) for your case studies (except for 30 Aug 2022, please add). Please compare these values to literature values, which might help to further constrain the aerosol type.

Hygroscopic growth parameters for different aerosols are summarized in the paper of Sicard et al. We added comparison with this paper.

Fluorescence quenching is a central topic of your manuscript. Please add some sentences to describe the phenomenon. In the introduction the term is mentioned, but not explained. A usual task for a reviewer is to ask for uncertainty estimates. I would appreciate if you put an uncertainty range to all values mentioned in the text.

Ln.50. This paragraph is modified as following:

“The addition of a fluorescence channel to Mie-Raman lidar (Veselovskii et al., 2020) opens new opportunities for particle characterization. If hygroscopic growth has no impact on aerosol fluorescence properties, fluorescence may serve as a proxy for evaluating the volume of dry aerosol. However, laboratory studies demonstrate that fluorescence can be suppressed ("quenched") by interactions with other molecules through processes of collision and energy transfer (Lakowicz, 2006), leading to reduction in emission intensity. In particular, water

molecules can efficiently quench fluorescence of organic fluorophores as reported by Dobretsov et al. (2014) and Maillard et al. (2021). Meanwhile, the extent to which such quenching occurs in atmospheric aerosols during hygroscopic growth remains an open question (Gast et al., 2024; Reichardt et al., 2025). Fluorescence quenching is expected to depend on aerosol composition, phase state, and presence of organic coating, highlighting the need to analyse a diverse set of measurement episodes.”

Minor comments:

I would prefer the term “surface area” and not just “surface”.

Changed

The date format is not consistent throughout the manuscript and the figures. I would prefer the order day month year as it is more logical than month day year.

Changed

Section 2 is written very concise. It would be helpful to add a sentence describing how the fluorescence backscatter is used to normalize the hygroscopic growth in case the particle number density changes with height. It is described in previous studies, e.g., Miri et al., AMT 2024, but it would be helpful for the general reader to include a short statement around L 123.

Eq.6 is expanded to explain normalization.

L 91-94 It is still a pity that you cannot launch radiosondes from Lille. Have you tried to use the (potential) temperature from GDAS instead the radiosonde from such a distance? Especially for the PBL, the variations to a far away radiosonde station might be tangible. Please discuss.

Yes, unfortunately RH measurements are not collocated. We used temperature profiles from both GDAS and radiosondes. Corresponding comment is added to revised manuscript.

Fig 1: Please explain SU and OC in the figure caption.

Done

And why are the model results just presented for a certain range of extinction coefficients?

Results are shown for 0-99% range of RH. We added Appendix with Table, containing OC and SU parameters used in computations.

L 179 Does the study provide an estimate of the uncertainty for urban aerosol in Paris? It would be helpful to justify the difference from the well-fitting value which you use.

Unfortunately, this study does not provide uncertainties.

L 218 The profile of the particle volume V^α should align with V^{3+2} because the conversion factor to derive V^α was estimated based on V^{3+2} . Is this correct?

Yes. The idea is to show that mean value of conversion factor can be applied to the whole profile to get volume with high height resolution.

L 323-324 From your results, I fully agree that reliable retrievals of the volume density should rely on the extinction coefficient. Please note that the method by Mamouri and Ansmann (2017) is based on the aerosol-type specific backscatter coefficient multiplied with the corresponding lidar ratio. In the case of hygroscopic growth as you present, this method will fail because it uses a fixed extinction-to-backscatter ratio disregarding the change of the lidar ratio due to hygroscopic growth. It was recently shown that the lidar ratio of urban aerosol increases with increasing RH.

Yes. We agree. The method of Mamouri and Ansmann (2017) works well in elevated smoke and dust layers, where hygroscopic growth is normally not observed.

Section 5: Actually, the conversion factors for the smoke particles are a bit off topic, because the main part of the manuscript deals with hygroscopic growth of urban aerosol. Nevertheless, I find it very interesting and relevant to include the conversion factors for smoke as well.

Extinction-to-surface-area conversion factors: Mamouri and Ansmann, ACP 2016, provides conversion factors for the surface area as well. The continental values of 2.8 (Germany, 532 nm) and 1.55 (Germany, 355 nm) agree well with your factors for urban aerosol.

We have added corresponding comment and reference to the manuscript.

Technical corrections

Some references are not correct, e.g., L 22 Burton et al., 2012 (according to your reference list) or L328 Fig 11 does not exist, it is Fig 10

Corrected

In the introduction and elsewhere, there are some fragments from a former version of the manuscript, e.g., L 29, L 63, L 269 which lead to incomplete sentences

Corrected.

Some abbreviations are not explained (but understandable): L 77 RI, L138 sigma

Corrected

L 162 Results of Observations

Corrected

L 176 On the second instance in this line, the indexing for the conversion parameter is done wrongly (alpha and V are swapped).

Corrected

L 247 Here you use a different unit for the WVMR

Corrected

L 229 and Fig 4d: the symbol for the real part of the refractive index is not consistent between text and figure.

Corrected

L 312: There is a smoke case from 2020 included in Fig 10., so it will be 2020 – 2024.

Corrected