Replies to Referee #2

We would like to sincerely thank the Referee for their careful reading of our manuscript and for their constructive and helpful comments. Their suggestions have helped us to significantly improve the clarity, structure, and scientific depth of the paper. In the revised version, we have addressed all comments point by point. Changes in the text are clearly marked in the manuscript *with red fonts*.
Below, we provide detailed responses (*red fonts*) to each reviewer's comment (*black fonts*) and indicate where the corresponding revisions have been implemented.

General comments:

I found this article very interesting. It presents a novel approach that uses lidar measurements to perform coarse pollen speciation in the lower troposphere.

The methodology is clearly explained, and the results are convincing. However, there is insufficient discussion of uncertainties. One important point is calibration. Certain aspects need to be clarified for readers so that they can rely on and use this work.

In my opinion, this article is entirely appropriate for ACP and can be published once the additional information requested has been provided. My corrections and questions are in the body of the article (in supplement).

This article provides a new perspective on the use of lidar technology and requires only minor revisions.

We thank the reviewer for the careful reading and helpful suggestions that improved the overall clarity and consistency of the manuscript.

We have defined all acronyms and terms at first use, added missing parentheses where appropriate, and corrected typographical and syntactic errors. Figure 1 has been updated to correctly position the "30°" label. In addition, many sentences that have already stated, have been omitted, and spacing inconsistencies have been corrected throughout the text.

Specific comments:

Comment:

If the density is lower, that is normal. Some information is missing.

Reply:

That's right. Corrected to: "During this campaign, pollen particles were detected near ground (up to 2 km height), showing strong fluorescence backscatter coefficients b_F at 355 nm (up to 8 x 10^{-4} $Mm^{-1}sr^{-1}$)." (L23-24)

Comment:

Could it also be related to dispersion during transport?

Reply:

Yes, that's right, the corrected paragraph is: "…. mixing with non-fluorescent particles and air mass dispersion processes." (L33)

Comment:

How many? (regarding the Temperature)

Reply:

Corrected to: "The latter can occur at relatively warm sub-zero temperatures (~ -2 to – 10 ºC), and thus bioaerosols can initiate ice multiplication that leads to rapid glaciation, storm intensification and extreme precipitation (Gao et al., 2025; Lohmann et al., 2016; O'Sullivan et al., 2015)."

Comment:

Dust and BB aerosols are not particularly organic.

Reply:

Agreed, the sentence was modified:" The main objective of this campaign was to understand the spatio-temporal variability of different types of bioaerosols containing pollen, BB and dust within the Planetary Boundary Layer (PBL) and lower free troposphere aloft (typically up to 2-5 km a.s.l.)"

Comment:

Could the uncertainty be higher by setting the LR? Different types of aerosol can have very different LRs. In addition to ageing, air masses can be mixed.

Reply:

Correct. Different aerosol types have different LRs, therefore, we performed a sensitivity analysis for the three aerosol types. Added to L232–233:
"The uncertainties of the retrieved b_{355} values, are of the order of 25-30%, based on a sensitivity analysis performed for these three different types of aerosols."

Comment:

The error becomes of secondary order because it is the ratio of two channels that is considered.

Reply:

Correct, thanks for noticing, added to L240: "…. the induced second order error on $\frac{T_E}{T_F}|_{par}$ for the particulate contribution remains less than 6% (Gast et al., 2025). "

Comment:

I understood that an elastic channel had been used. Could you confirm this?

Reply:

Yes, we confirm. The paragraph was modified (L247–251): “Furthermore, to equalize (calibrate) the PMT sensitivities, we installed the PMT from the fluorescence channel to the so called “Raman” one (Veselovskii et al., 2020); then, by adjusting the voltage supply, we obtained the same signal intensity P_{F355} as the elastic one P_{E355} at the analog channel at 355 nm. This equalization (calibration) ratio can be expressed by the ratio $[\frac{P_{E355}}{P_{F355}}]_{an,cal}$ both at 355 nm, along the whole range of the analog channel and is found to be $[\frac{P_{E355}}{P_{F355}}]_{an,cal} = 0.12$.”

Comment:

What level of uncertainty is associated with such a calibration? Can the calibration of the elastic channel be verified over an altitude range where molecular scattering dominates?

Reply:

We only measured in altitudes where bioaerosols dominated. Revised to (L256-257) hus we transformed the sentence into:

“Thus, in our LIF lidar system we acquired a value of F=65 when gluing the P_{Fpc} (in MHz) to P_{Fan} (in mV) within the altitude range where the bioaerosols dominate.”

Comment:

What is the deconvolution function? It's not clear.

Reply:

Apologies for the confusion, its actually the method of spectral decomposition. Thus, we transformed the L266-267: “The method is based on the spectral decomposition of the LIF lidar signals obtained by a multichannel lidar detector to determine the contribution from each taxon.”

Comment:

I am wondering whether this is really a deconvolution in the mathematical sense of the term, or whether it is proximity recognition using a cost function?

Reply:

Please see the above response. Please see the clarification above — it is a spectral decomposition method, not a mathematical deconvolution.

Comment:

The introduction does not address health issues; perhaps a few lines should be added.

Reply:

Thank you for noticing, we added a few lines in Introduction regarding health issues: “This prolonged exposure has been linked to a rise in allergic respiratory diseases such as allergic rhinitis and asthma, affecting millions of people worldwide and imposing a growing public health burden, especially in urban environments where interactions with air pollutants can further intensify symptoms (Buters et al., 2018; D’Amato et al., 2020). Understanding the

dynamics of pollen emission, transport, and transformation in the atmosphere is thus crucial for improving allergy forecasts and assessing health risks under changing climatic conditions." (L53-57).

Comment:

This paragraph should be separated between the introduction and Section 2.

Comment:

A subsection titled 'Pollen climatology of the site' would be included before 4.1.

Reply:

Implemented as suggested. A new subsection titled *Pollen Climatology of the Site* has been added before Section 4.1.

Comment:

Do you use Hysplit in ensemble mode? Over a period of 10 days, the cumulative errors are significant on the back trajectories, and it is preferable to use multiple runs.

Reply:

Thank you for the suggestion, we added it in the Supplement.

Comment:

Temperature is one factor to consider, but shouldn't we also pay attention to the importance of solar radiation?

Reply:

Correct. Added to L328-329: "It is well documented that grass pollen concentration levels generally peak during daytime (generally higher solar irradiance and temperature levels compared to nighttime) and often tend to increase after rainfall (Kelly et al., 2013; Sabo et al., 2015)."

Comment:

These two meteorological variables are generally anticorrelated.

Reply:

We agree. Many field studies (including CALISHTO at Mt Helmos) observed anticorrelations between T and RH, although situations with high T and low RH can still occur.

Comment:

What are the error bars on these measurements?

Reply:

The metrological quantification of both manual and automatic measurement is an ongoing effort (https://www.bioairmet.ptb.de/). We are not yet able to provide precise quantification of the errors.

Comment:

Pollen is generally not included in PM2.5 or even PM10 measurements. How can this data be linked to pollen concentrations?

Reply:

Particles smaller than 10 micrometers are filtered out after the trigger laser stage in the Swisens Poleno. Conversely, Pollen grains are too large to be included in PM10 measurements. Those measurements do not overlap.

Comment:

Could you elaborate on the significance of ion chromatography in this study?

Reply:

Ion chromatography is used to identify and quantify major inorganic ions, providing chemical context that helps distinguish aerosol sources (e.g., biomass burning, pollution, or secondary formation) relevant to interpreting lidar observations.

Comment:

For the first case study, there is no INP. It may be better to introduce the differences between the two case studies.

Reply:

INP measurements were not available for 27–30 May. The two days with INP data are 24 and 31 May, and this distinction has been clarified in the text.

Comment:

Where would they come from?

Reply:

Coarse-sized particles could be soil dust and pollen, or larger sized fungal spores originating from local forest and agriculture lands. We do not have Dust event on these days. Added in L484-485: "… which suggests coarse-sized particles, such as soil dust, pollen and large-sized fungal spores contribute to the observed INPs at –20°C."

Comment:

Is the consideration of RH related to the hygroscopicity of the particles and the associated decrease in fluorescence?

Reply:

Correct. Added to L582-585: "Along this transport the b_F values passed from ~4.5 x 10^{-4} $Mm^{-1}sr^{-1}$ to 2.0 x 10^{-4} $Mm^{-1}sr^{-1}$, now showing a ~50% reduction on the b_F values, indicating again a possible mixing with non-fluorescent (e.g. continental polluted) aerosols of different hygroscopicity values at lower heights."

Comment:

It should be emphasized in the abstract that this is a new approach which reliably provides information on pollen speciation in the lower troposphere.

Comment:

Always in the abstract, it is fair to say that this is an original approach.

Reply:

Thank you for noticing. Added to L21-L23: “This original approach provides, for the first time in this region, reliable information on pollen speciation aloft, bridging the gap between ground-based sampling and remote sensing observations.”