Response to Referee #1

Thank you for carefully reading the manuscript and providing useful suggestions to improve the paper. The replies to the referee comments are given below. The referee comments are highlighted in blue with our responses in black. The sentences in the manuscript are in Italic, with the modifications in the revised manuscript in red.

The authors describe a methodology to infer the particle linear depolarisation ratio (PLDR) of different types of airborne pollen in their pure state, i.e. unmixed with ambient aerosols. Due to the size and irregular shape of pollen, it can be expected that their PLDR is larger than that of most aerosol types though likely smaller that that of mineral dust. Knowing the PLDR of undiluted aerosol types is important as it allows for separating, in the presented cases, the contribution of pollen to pollen-containing aerosol mixtures.
I am not quite sure that the authors are actually presenting simulations or modelling in their discussion of a simulator. It all seems quite analytical.
We are presenting a simulation in this paper. In the simulator section, we applied the algorithm to the synthetic lidar data. This part will be clarified in the revised version.

In the revised manuscript, Page 6 lines 4-6 will be:

This algorithm is first tested through a simulator (Sect. 3.3.2) using the synthetic lidar data, and then applied to the real lidar observations (Sect. 3.4). The simulator includes a direct model and an inverse model modules (the block diagram is shown in Fig. S1 in the supplement); Similar ones have already been used for forest and aerosol studies (Shang et al., 2018; Shang and Chazette, 2015). Synthetic data are used in this section to present our methodology.

To my understanding, the shape of the extinction coefficient profiles for pollen and the background aerosol is defined as given in Table 3. The magnitude of the extinction coefficient of both species is then determined by the respective optical thickness which is also set. Backscatter coefficients are obtained using the set lidar ratio and the Angstrom exponent is derived trough mixing the set values of the two aerosol types.
Yes, this is the way we simulate the extinction and backscattering coefficients.

It is totally unclear, though, how the authors arrive at the mixed PLDR profiles presented in Figs. 4 and S2b. PLDR values for both types are also set in Table 3 but the mixing rule is not linear such as summing up the backscatter or extinction coefficients. Is this when simulations come into play? If so, what is done to get the PLDR profiles? It might be that the authors have used Eq. (6) which is given in Section 3.3.2 to obtain PLDR profiles of the pollen-background-aerosol mixture. If so, the entire simulator would be circular as identical calculations would be done in both directions. This would explain the perfectly linear relationships presented in Figs. 5, 6, and 7. I am afraid I cannot assess the scientific quality of this work before the authors clarify the description of the simulator, particularly with respect to the points made above.
We will clarify the PDR calculation in the revised version. The calculation is as following:

We follow the detailed calculations in Tesche et al. 2009.
The particle depolarization ratio ($\delta_{particle}$) is expressed as Equation 4 in the manuscript:

$$\delta_{particle} = \frac{\beta^{\perp}_{pollen}+\beta^{\perp}_{background}}{\beta^{\parallel}_{pollen}+\beta^{\parallel}_{background}}, \qquad (4)$$

The depolarization ratio of one particle type can be defined as:

$$\delta_x = \frac{\beta^{\perp}_{x}}{\beta^{\parallel}_{x}}, \qquad (S1)$$

The index *x=pollen* or *background* denotes the contribution of pollen or background particles, respectively. We can use the following relationships mathematically:

$$\beta_x = \beta_x^{\perp} + \beta_x^{/\!/}\ , \tag{S2}$$

$$\beta_x^{/\!/} = \frac{\beta_x}{1+\delta_x}\ , \tag{S3}$$

$$\beta_x^{\perp} = \frac{\beta_x \delta_x}{1+\delta_x}\ , \tag{S4}$$

We replace equations S3 and S4 in equation 4, the particle depolarization ratio can be then calculated using the particle backscatter coefficients (β_{pollen} and $\beta_{background}$) and the depolarization ratios of both particle types (δ_{pollen} and $\delta_{background}$):

$$\delta_{particle} = \frac{\frac{\beta_{pollen} * \delta_{pollen}}{\delta_{pollen}+1} + \frac{\beta_{background} * \delta_{background}}{\delta_{background}+1}}{\frac{\beta_{pollen}}{\delta_{pollen}+1} + \frac{\beta_{background}}{\delta_{background}+1}}\ , \tag{S5}$$

These equations and descriptions will be added in the supplement for the revised version.
In the revised manuscript, Page 6 lines 24-26 will be:

Next, pollen layer and background layer are summed up, and then the vertical profiles of aerosol backscatter coefficient, ~~particle depolarization ratio,~~ lidar ratio and Ångström exponent of the total aerosols are simulated (e.g., Fig. S2b); Vertical profiles of particle depolarization ratio can be also calculated following eq.S5 in the supplement (the detailed calculation is also given). Theoretically, these parameters can be derived directly from lidar observations.

Further comments

• The title is a bit misleading as the reader might expect observations from an aircraft. I'd suggest a clearer title such as "Optical characterisation of airborne pollen from lidar measurements in Finland"

Thank you for the suggestion, we will change the title as:

Optical characterization of pure pollen types using a multi-wavelength Raman polarization lidar.

We have also removed "in Finland" as the presented method can also be applied to other sites.

• The part about CALIPSO in the Introduction (page 2, lines 21-28) should be omitted. It is not needed as there is no later reference on how to apply the new results of this work to improve the CALIPSO aerosol typing.

We agree. We will remove the related information in the introduction for the revised version.

• page 3, lines 3-13 are more suitable in the introduction

We agree that these sentences can also be placed in the introduction, but we think these sentences are more suitable for the description of our campaign site. So we decide to keep them in the section 2.

• I'd suggest a change of the structure of the paper: 1. Introduction, 2. Site and instruments, 3. Lidar simulator, 4. Results. Such a structure allows for a clear separation of instruments, methods, and findings. In addition, the lidar parameters should already be introduced in the description of the lidar. This way, the reader knows what's available for the theoretical studies in the next section.

Thank you for the suggestion, we were considering the suggested structure, which is also good. Still we decided to keep the current structure as we find it logical for the purpose.

In the section 3 Methodology and results, we present first the results from Burkard sampler (Sect. 3.1), based on which we define the intense pollination periods. Secondly, we present the optical properties of the pollen layer (Sect. 3.2). Which are a quasi-straightforward results and can be easily retrieved from lidar observations. Thirdly, we present the optical properties of pure pollen (Sect. 3.3 and 3.4), first by the synthetic simulation then applied to the lidar measurements. In this third part, we introduce the algorithm along with the assumptions.

The optical properties retrieved from lidar were presented in section 2, page 3 lines 16-24.
We will add more descriptions in section 2 as (page 3 lines 16-24):

Polly^{XT} has three emission wavelengths (355, 532 and 1064 nm) and seven detection channels (including three emitted wavelengths channels, three inelastic Raman-shifted wavelengths channels (387, 407 and 607 nm) and the cross-polarization channel at 532 nm). During daytime, the Klett-Fernald method (Fernald, 1984; Klett, 1981) is applied using the elastic signals to retrieve the extinction coefficient which describes the combined effect of particle absorption and scattering, and the backscatter coefficient which describes particle backscattering at 180° scattering angle. During night-time, profiles of extinction and backscatter coefficients at 355 and 532 nm can be derived independently using elastic and inelastic Raman-shifted wavelengths (387 and 607 nm), based on the Raman inversion (Ansmann et al., 1992). The ratio of extinction to backscatter coefficient is called lidar ratio (LR), which is considered an important parameter to separate particle types, as it depends on their single scattering albedo and backscatter phase function, thus being a function of size distribution and chemical composition. The cross- and total- polarization channels of the Polly^{XT} allow the retrieval of the volume depolarization ratio (VDR) and linear particle depolarization ratio (PDR) at 532 nm, which provide information on the shape of the scattering particles. Multi-wavelength measurements (355 nm, 532 nm and 1064 nm) enable the determination of Ångström exponents between each wavelength pairs, which are related to the particle nature, mostly the size. The operated lidar system has an initial spatial resolution of 30 m and a temporal resolution of 30 s.

• Are there any objective criteria for determining the exact times of the different intensive pollination periods? I am thinking of a certain threshold of the extinction coefficient at a certain height or similar quantifiable criteria.

In this study, we mainly considered the pollen concentration measured by Burkard, the daily mean pollen concentrations were used as a constraint. We will add this information in the revised version as (page 4 lines 24-25):

Four intense pollination periods (IPPs) are defined considering ~~both~~ the pollen seasons and the daily mean pollen concentration values of these 4 dominant pollen types (Table 1). A minimum value of 300 no. m^{-3} were applied for birch and pine pollen for IPP-1 and IPP-3, whereas a smaller value of 20 no. m^{-3} were applied for spruce and nettle pollen for IPP-2 and IPP-4. In addition, the availability of ~~available~~ lidar measurements were considered for the IPP definition.

This method is good enough for our measurement site, and can be used for the other sites under the condition that there is no other depolarization particles. For some sites, more criterions would be needed to define a good pollination period, for example using back-trajectory or in situ measurements to avoid dust effect periods/layers. In this study, we considered the 1st aerosol layer near ground using the layer definition method.

• It would be good to get some information on how often pollen are observed at such high altitudes and how they get up there in the first place.

That's a very good point. And it is included in our future work plan. In our other campaigns, we used the drone measurements for such study. But this part is not in the scope of the presented study.

We have explained the long distance transport of pollen in our previous paper (last paragraph of section 4.1 in Bohlmann et al. 2019), and we will add the sentence in the revised version as (Page 2, line 9):

Several studies on the long distance transport of pollen (Rousseau et al., 2008; Skjøth et al., 2007; Szczepanek et al., 2017) have shown that pollen grains can be lifted up to several kilometers and be dispersed by wind over thousands of kilometers.

- **The abbreviation PBC is not ideal as it is often used to denote the particle backscatter coefficient. I'd suggest to rename this into some ratio with a different variable.**

Thank you for point this out! We agree, and we will change this PBC to χ_{pollen} for the whole manuscript (including figures).

For example in Page 6 lines 29-31:

Pollen backscatter contribution ~~(PBC)~~ inside the pollen layer from heights z1 to z2 (in this simulation z1 = 0, z2 = 1 km), denoted as χ_{pollen}, is defined as the ratio of pollen backscatter coefficient (β_{pollen}) and the total particle backscatter coefficient ($\beta_{particle}$). Note that the use of "particle" here is to distinguish from "molecular".

$$\chi_{pollen}(z1, z2) = \frac{\int_{z1}^{z2} \beta_{pollen}}{\int_{z1}^{z2} \beta_{particle}}, \tag{1}$$

- **There is no mentioning of the assumed shape of the pollen and background aerosol profiles in the uncertainty study. Is there any justification for selecting this shape? How general are they? Is the same pollen profile assumed in the inverse model?**

There is no assumption of profiles for the inverse model, as it uses the output of direct model as its input.
We used the same shape of profiles as presented in section 3.3.1. In the section 3.3.3 (page 8, Line 25), we mentioned "*using the parameters of previous simulated 6 cases (Sect. 3.3.1)*".

We agree that it was not very clear. We will clarify this in the revised version as (Page 8 line 25):

The uncertainty study of this method is investigated in this section~~, using the parameters of previous simulated 6 cases (Sect. 3.3.1)~~. The input parameters of the direct model are defined in 3.3.1, with optical depth (OD) of the background aerosol of 0.1, and pollen OD of 0.002, 0.01, 0.02, 0.05, 0.1, or 1. Nonetheless, some input parameters (e.g., the pollen depolarization ratio δ_{pollen} and the backscatter-related Ångström exponent for pollen $\hat{\text{Å}}_{pollen}$) were selected as different values for different uncertainty studies, which are clarified in each paragraph. The output of each direct model simulation were then used as the input of the inverse model.

Also in Page 9 line 10 will be modified as:

The parameters for the 6 cases simulated earlier ~~previous simulated 6 cases~~ (as defined in Sect.3.3.1, with values given in Table 3) are used again ~~used~~ in this simulation

- **Why is the pollen layer set to extend from 0 to 1 km when the measurements (1) only start at 600 m or so and (2) show pollen all the way to 2 km height?'**

This is the simulation of synthetic lidar data, the results don't change if the assumed layer heights change. In the simulation we assumed a pollen concentration with a layer center of 0.5km and half width of 1km. This is why we defined the pollen layer as 0-1 km. Theoretically, we can change the input values of the direct model as whichever reasonable values; in the given example, we just simulate one case. It is also possible to simulate a case with pollen concentration from 600m to ~2km, as the ones from the real measurement pollen layer. But the conclusion of the simulation section would remain the same.

We will clarify this in the revised version as (Page 6, line 17):

In addition, the conclusion of the simulation section is not depended on the assumed profile shape or height.

• It would also be good to get an idea of typical values of total and pollen-related optical thickness at your site to assess the choice of values in your method. Is a pollen optical depth of unity even possible?

This is a good point, and we are working on it. We are collecting more data, so as to provide more statistic values.