Response to Referee #3

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 between the quotation marks, with the modifications in the revised manuscript in red.

The authors present multi-wavelength Raman polarization lidar measurements of pollen layers in Finland combined with a Burkard pollen sampler. Active remote sensing measurements of pollen are rarely found in literature. Therefore, the present manuscript enriches our knowledge about the optical properties of abundant pollen types such as birch and pine pollen. Northern Europe (Finland) is a good location for such a study as it is less affected by other depolarizing aerosol particles such as mineral dust. Additionally, the authors present a novel approach to derive the depolarization ratio of pure pollen layers. Although it is related to some uncertainties, it is a big step forward compared to just presenting the layer mean values. I support the idea that measurements of the depolarization ratio at various wavelengths should be enforced in future pollen-related studies. Polarization lidars may in future support pollen forecasts and help citizens with pollen allergy thanks to the characterization of pure pollen types by these authors. The quality of the figures and tables is high.
Finally, I recommend publication after minor revisions.

Major remarks:

1. You use a value of 3 for the backscatter-related Ångström exponent of the background aerosol. Do you have any statistical evidence of this value for the station at Kuopio? Is it a mean value for the pollen-free periods? And how sensitive is your analysis to this assumption?

Thank you for pointing it out. The choice of parameters in Table 3 (Table 1 in the revised version) for the simulation is not critical for presenting the overall approach.
We have changed the assumption value for non-pollen particle Ångtröm exponent ($\mathrm{Å}_{\mathrm{background}}$) as 2 (instead of 3) in the revised version. This value of 2 is more realistic. We have changed all the related results and figures. The assumption for $\mathrm{Å}_{\mathrm{background}}$ is only used in the simulation part, and is not considered for the pollen depolarization ratio retrieval, so the actual results using lidar measurements will not change. We have made some modifications for the revised version to make is more clear.

We have added information in section 3.1 as:
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> The optical and physical parameters used in the direct calculation are presented in Table 1; these parameters are named as "initial values" for the simulation. The values are based on our lidar measurements (Bohlmann et al., 2019) or literature (e.g. Illingworth et al., 2015). The *background* here refers to non-depolarizing background aerosols (non-pollen particles), which can be polluted continental or biomass burning aerosols. The depolarization ratio at both 355 and 532 nm of non-pollen particle ($\delta_{\mathrm{background}}$) are selected as 0.03, which is a mean value for pollen-free periods at our measurement site. Bohlmann et al. (2019) shows that the pollen can generate strong depolarization, thus the depolarization ratio at 532 nm of pure pollen particle (δ_{pollen}) are selected as 0.35 as the initial value for the simulation in this section. Pollen grains are quite big and thus can be assumed to be wavelength independent on the backscatter at wavelengths of 355 nm and 532 nm, with the backscatter-related Ångström exponent ($\mathrm{Å}_{\mathrm{pollen}}$) of 0. The backscatter-related Ångström exponent between 355 and 532 nm of non-pollen particle ($\mathrm{Å}_{\mathrm{background}}$) is assumed to be 2, regarding the previous studies over Arctic regions (e.g. Schmeisser et al., 2018; Tomasi et al., 2012). Note that these values can be changed freely for the simulation under 2 constraints: i. depolarization ratio of pollen (depolarizing one) should be higher than the depolarization ratio of background aerosol (non-

depolarizing one), ii. the values of backscatter-related Ångström exponent for pollen and non-pollen particle should be different. In addition, the conclusion of the simulation section is not depended on the assumed profile shape or height; and the initial values are not critical for presenting the overall approach.
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We have also investigated the sensitivity of this assumption in the simulation section. For the uncertainty study due to initial and assumed Ångström exponent in section 3.3 of the revised version, we have modified as:
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In the presented cases, we assumed that the backscatter-related Ångström exponent between 355 and 532 nm of pure pollen to be used in the inverse model (denoted as $\widehat{\text{Å}}_{\text{pollen}}$) is 0, which was the same as the initial value (Å_{pollen}) of direct model. But in the reality, such information is not always available. Under different initial values of Å_{pollen}, there will be a bias on the estimated values of pollen depolarization ratio if the assumed value is different (i.e. $\widehat{\text{Å}}_{\text{pollen}} \neq \text{Å}_{\text{pollen}}$). For example, if the initial value Å_{pollen} is 0.25 (i.e. η_{pure}=1.11), but we keep the assumption of $\widehat{\text{Å}}_{\text{pollen}}$=0 in the inverse model, the estimated pollen depolarization ratio is found to be 0.39 with a bias of 0.04 (show in Fig. S3 in the supplement). The uncertainty due to the difference between the initial value of Å_{pollen} and assumed $\widehat{\text{Å}}_{\text{pollen}}$ were simulated (show in Fig. S4 in the supplement), where $\widehat{\text{Å}}_{\text{pollen}}$ is always assumed as 0 in the inverse model. For initial values of Å_{pollen}=±0.5 (i.e. bias of 0.5 on the assumed value of 0), relative uncertainties were assessed as ~30 %. This uncertainty due to the difference of initial values of Å_{pollen} and $\text{Å}_{\text{background}}$ was also investigated. The larger the difference between two values ($\text{Å}_{\text{background}} - \text{Å}_{\text{pollen}}$), the smaller the uncertainty. For instance, if we use 3 (instead of 2) as the initial value of $\text{Å}_{\text{background}}$, the estimated pollen depolarization ratio is 0.37 (instead of 0.39) with a smaller bias for the above example.
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In addition, we can retrieve the non-pollen particle Ångtröm exponent using our lidar measurements, based on the presented algorithm (using Eq.5 in the revised version). We found $\text{Å}_{\text{background}}$ values of 2.0 and 1.9 for IPP-1 and IPP-3, respectively. These results are added in section 4.3.1 in the revised version:
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Under the assumption that the backscatter-related Ångström exponent between 355 and 532 nm of pure pollen (denoted as Å_{pollen}) is 0 (i.e. η_{pure}=1), depolarization ratio of 0.24 or 0.36 were found for IPP-1 or IPP-3, respectively, which are related to the pure birch or pure pine pollen (Table 5). The scatter plots of mean η and $\chi_{\text{pollen}}(\delta_x, 532)$ are shown in Fig. 12: (a) for IPP-1 with the pollen depolarization ratio of 0.24, and (b) for IPP-3 with the pollen depolarization ratio of 0.36. Good linear regression relationships are found for both cases, and two things should be highlighted: (1) Å_{pollen} is 0 (i.e. η_{pure}=1) for 100 % pollen in the observed aerosol particle population (i.e. χ_{pollen}=1); (2) without pollen in the air (i.e. χ_{pollen}=0), the backscatter-related Ångström exponent between 355 and 532 nm of non-pollen particles ($\text{Å}_{\text{background}}$) can be calculated, resulting values of 2.0 for IPP-1 and 1.9 for IPP-3 (i.e. η of 2.28 for IPP-1, 2.18 for IPP-3).

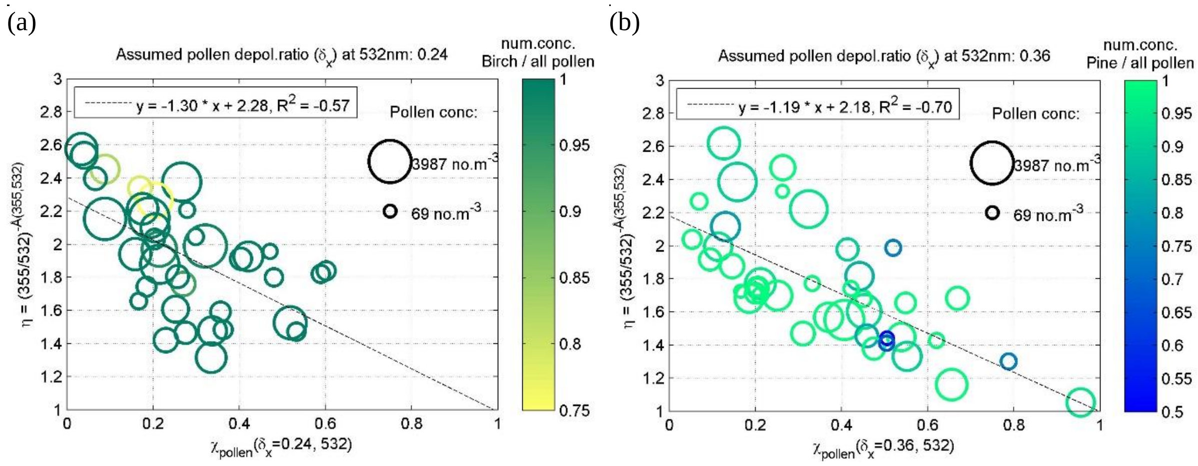

Figure 12. Mean values of the parameter η against pollen backscatter contribution at 532 nm ($\chi_{\text{pollen}}(\delta_x, 532)$) inside the pollen layers, during the IPP-1 (a) and IPP-3 (b). η is a parameter using backscatter-related Ångström exponent between 355 and 532 nm (Eq.6). The pollen depolarization ratio δ_x at 532 nm is assumed to be 0.24 for (a) or 0.36 for (b). Linear regression lines are drawn by dotted lines, with fitting equation shown (Eq.5 or 8). The correlation coefficient (R^2) is also given. The size denotes the total pollen concentrations measured by the Burkard sampler on roof level; the colour represents the number concentration of the dominant pollen (a: birch, b: pine) against the total pollen number concentration. Similar figures using different assumed values of pollen depolarization ratio can be found in Fig. S5 and Fig. S6 in the supplement.

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2. Your novel approach for getting the depolarization ratio of the pure aerosol type is remarkable. I am just wondering whether the mixture of continental background aerosol and pollen has a significant effect on the lidar ratio, too. It would be great to have the lidar ratio and the depolarization ratio for pure birch and pine pollen at the end. Please comment on this.

We think the pollen has effect on the lidar ratio. But more nighttime measurements (for lidar ratio retrieval) for intense pollination cases are need for investigating such scientific question. If we find a case where there is a pollen layer in the free troposphere (without contamination of aerosols in PBL), with strong depolarization ratio and small Ångström exponent, it will be good to study the lidar ratio for such layer to retrieve “pure values”.

Minor remarks:

3. P5,L25: “The extinction-related and backscatter-related Ångstrom exponent were also retrieved for pollen layers.” – Is the extinction-related Ångström exponent shown somewhere? It must not be shown in the manuscript, some descriptive words are sufficient.

We have retrieved the extinction-related Ångström exponent, but we haven’t presented such parameter as the available data are limited. We have added descriptions as:

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> The extinction-related (not shown in this study) and backscatter-related Ångström exponent were also retrieved for pollen layers.

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4. P10,L30 The Ångström exponent is related to extinction or backscatter?

Thank you for pointing this out. It is the extinction-related, we have added information in the revised version.

“

> For big particles as dust, Mamouri and Ansmann (2014) reported extinction-related Ångström exponent between 440 and 675 nm with values of -0.2 for coarse dust and 0.25 for total dust.

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5. P11,L10 Are the measurements presented by Cao et al., (2010) performed at exactly 180_ backscatter direction? This is not so easy to achieve in chamber experiments. Maybe there is an additional source for the discrepancy arising from the optical design of the Cao measurements?

Cao et al. (2010) performed the measurement at 180 deg direction. The lidar measurements were made in an aerosol chamber located 100 m away from the lidar. 2 g of the selected pollen is disseminated within a few seconds with a pneumatic nozzle in the chamber.

They have pointed out that "the reported values are not exempt from specificities regarding the experiments as they were conducted" and have a discussion on this aspect. For example, the dissemination device used has an influence on the amount of agglomeration of the particles, and that certainly could affect the depolarization ratios. RH can be another reason, as for their experiment dry aerosols are being dispersed, whereas in our study, we focus on the aerosols in the atmosphere.

As we mentioned in the manuscript:

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> These values are higher than what we retrieved in this study, but it has to be kept in mind that these two experiments have been conducted in quite different environments and conditions.

"

6. Fig. 1+2 and Tab. 1: Please provide the year (2016) whenever you provide dates. Do it in the caption or just like this "Date mm/dd in 2016 [UTC]".

Thank you for your comment. The corrections have been done.

7. How do you get to the uncertainty range +/-5% for pine pollen? Varying the Ångström exponent by +/- 0.5 leads to values of 26 to 44% (Fig. 12 and P11,L9).

Thank you for pointing it out, we agree that it was confusion and not correct. We made the corrections to make it more clear in the revised version. Please also check our reply to the comment "Major remarks 1." for the uncertainty study for the simulation section.

For the uncertainty study of the real lidar measurements, we modified as:

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> Uncertainty study was investigated based on method describe in Sect.3.3 using a Monte Carlo approach. The overall relative uncertainties of the lidar-derived backscatter coefficients are of the order of 5 %–10 % (Baars et al., 2012), we took 10 % here in the simulation. Initial pollen depolarization ratio values were selected as 0.24 for birch and 0.36 for pine for the uncertainty simulation; initial backscatter-related Ångström exponent between 355 and 532 nm of non-pollen particles were selected as 2.0 and 1.9 for IPP-1 and IPP-3, respectively. Based on the lidar observations (Fig. 12), the simulated cases were selected so that the χ_{pollen} values range from 2 % to 60 % for birch and 2 % to 90 % for pine. The initial input $\text{Å}_{\mathrm{pollen}}$ in the direct model and assumed $\widehat{\text{Å}}_{\mathrm{pollen}}$ in the inverse mode were both selected as 0. Estimated uncertainties were found as 2.4 % for birch and 2.9 % for pine (Table 5). Note that the different initial input values of $\text{Å}_{\mathrm{pollen}}$ may introduce important additional bias. If we assume the true value of $\text{Å}_{\mathrm{pollen}}$ is between -0.5 to 0.5 (i.e. values of η_{pure} from 0.82 to 1.22, shown by red dotted lines in Fig. 11), depolarization ratios of 0.19 to 0.27 can be found for birch pollen, and 0.26 to 0.44 can be found for pine pollen.
>
> Table 5. Linear depolarization ratios for pure pollen. The assumption of backscatter-related Ångström exponent between 355 and 532 nm for pollen should be 0 was applied for this study. The uncertainty on backscatter-related Ångström exponent of pollen was not taken into account for the standard deviation shown here, which may introduce non-negligible additional bias. See more details in Sect. 4.3.

	Pollen type	Depolarization ratio at 532 nm	Depolarization ratio at 355 nm
This study, Finland	Silver birch	0.24 ± 0.01	0.17
(in the atmosphere)	Scots pine	0.36 ± 0.01	0.30
Cao et al. (2010), Canada	Paper birch	0.33 ± 0.004	0.08 ± 0.008
(in an aerosol chamber)	Virginia pine	0.41 ± 0.006	0.20 ± 0.013

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And in the conclusion we modified as:

"

> This algorithm was first tested and validated through a simulator of synthetic lidar profiles (including a direct model and an inverse model modules). Mathematically, the depolarization ratio for pure pollen can be calculated using the equations given in Sect. 3, if other variables are known or can be assumed. We have developed a retrieval method to estimate the pollen depolarization ratio, which was applied to the lidar observations. The depolarization ratio at 532 nm of pure pollen particles was assessed, resulting to 0.24 ± 0.01 and 0.36 ± 0.01 for birch and pine pollen, respectively. The uncertainty on assumed backscatter-related Ångström exponent of pure pollen will introduce non-negligible bias in addition as discussed in Sect. 4.3.1.

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Technical remarks

- Affiliations: "P.O. Box 1627, 5 70211" – seems not necessary and isn't provided for the other institutes

The correction has been done.

- P1,L11 / P2,L32: depolarization ratio values/value

The correction has been done.

- P3,L17: volume linear depolarization ratio (VDR) and particle linear depolarization ratio (PDR)

The correction has been done.

- P4,L18: spoken communication – with whom? Please acknowledge the name of the person

Thank you for your comment. We have added such information as:

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> *B. pubescens* pollen grains are 18-24 × 22-28 µm in size (Nilsson et al., 1977) and *B. pendula* (Silver birch) pollen grains are more or less of the same size (spoken communication with Sanna Pätsi from Aerobiology, University of Turku).

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- P6,L10: non-depolarizing aerosol – the received light is depolarized, but the aerosol is depolarizing, please change it throughout the manuscript

Thank you for pointing it out, we have modified it throughout the manuscript.

- P6,L12+L30: this type of indices should not be written in italic – please change it throughout the manuscript

We have changed these indices to non-italic.

- P6,L21: "thus six pollen backscattering are simulated." – backscatter coefficients or backscatter coefficient profiles (similar P12,L8)

Thank you for pointing it out, the correction has been done.

- P9,L8/9: It would be a good idea to begin a new paragraph with line 9

We agree. Actually it was a new paragraph, but it was not shown with the presented format.

- Fig. 1, caption of y-axis: [no m-3] – it is -3

- Fig. 3a, caption of y-axis: LR 532 [sr] – unit is missing

Thank you for pointing them out. The corrections have been done.