

Reply to Dr. Holger Baars

Thank you for your comments! Below you find our detailed response to each question.

Methodology:

1) If you speak about concentration of Pollen you refer to the number concentration, but what does this mean in terms of the surface concentration and the volume concentration which are the important parameters for backscatter and extinction of light? Can you clarify with typical sizes from the detected Pollen?

Thank you for this comment! Typical sizes of pollen are given in the results section. Also a comment about the volume concentration was added when explaining the effect of spruce pollen on the measured values despite the low number concentration contribution, as:

The Ångström exponent in the second period is around 0.8, whereas it is around 1 in the first period, demonstrating the effect of the larger spruce pollen (~90-110 μm), even with a small contribution (~14%) to the total pollen number concentration. But considering the different volume of birch and spruce pollen grains, the contribution of spruce to the total volume concentration exceeds 75% in the second period, which explains the large effect of spruce pollen on the measured optical properties even with a small number concentration.

2) Furthermore, can you provide a table with the optical and microphysical properties of the different Pollen types you analyse as detected in the laboratory, e.g. the depolarization ratio (Cao et al.), average density of Pollen, refractive index, and fall velocity (would be interesting as most Pollen have good flight behaviour.)

Thank you, this is a good suggestion, but optical and microphysical properties of both of the pollen types are either not reported in literature yet or vary significantly as for example the hydration of pollen grains varies and affects the density and therefore the fall velocity. A comment about the fall velocity of birch pollen was added in the result section, but because spruce pollen are much less studied and to our knowledge there are no laboratory measurements of optical properties of spruce pollen yet, we prefer not to provide a table as it would be incomplete and it is not crucial for the understanding of our study.

3) A discussion on the flight behaviour would be beneficial to relate the lidar measurements to the surface measurements. One could imagine that if particles are too large and too heavy and have thus a high fall velocity they would not make it up to the heights of the lidar observations (this could, e.g. another reason for your changing particle depolarization with height: Is the size distribution of the observed Pollen types nearly one size or can they change?).

Airborne pollen are designed in a way so that the grains can be dispersed by wind. Bigger and thus heavier pollen grains possess air bladders, which increase the surface area of the grain without adding much mass. This increases the amount of drag and decrease the settling velocity of the pollen grains. In this way it is also possible for big pollen grains to be lifted and dispersed by wind. In various studies e.g. Skjøth, 2007; Rousseau, 2008; Szczepanek, 2017, the long range transport of pollen has been shown. The size distribution of pollen within one pollen type is usually narrow. The size varies only little within one specie (Mäkelä, 1996; Kishchenko and Tikhova, 2019). A comment about the flight behavior of pollen is added in section 4.1., as:

Pollen are low density particles, which makes them more sensitive to air currents, reduces the settling velocity and allows them to be lifted by turbulent air flows. Birch pollen, for example, have a gravitational settling velocity of around 1 cm s^{-1} (Sofiev et al., 2006). This settling velocity is similar to anthropogenic aerosol smaller than $10 \mu\text{m}$ (PM10) although birch pollen grains are more than twice the size. The air bladders on the bigger spruce pollen grains increase the surface area of the grain without adding much mass, and therefore decrease the settling velocity. Hence, even those big pollen grains can be lifted up to several kilometers and be dispersed by wind over thousands of kilometers as have been shown by several studies on the long distance transport of pollen (Rousseau et al., 2008; Skjøth et al., 2007; Szczepanek et al., 2017).

4) You state that during your observations the lidar ratio of Pollen stays the same while depolarization changes. What theoretical optical properties do you expect from, e.g., scattering calculations? Is there any literature available? Can you explain how the depolarization of the laser light is working at this size range?

Unfortunately there are no theoretical optical properties for the observed pollen types under ambient conditions, e.g. hydrated pollen. The paper by Cao et al. provides experimental values for various pollen types but doesn't have any values for example for spruce pollen. Furthermore dry pollen grains were used which have different optical properties. We will try to obtain scattering simulations for future studies as theoretical reference values for the optical properties of different pollen types under different environmental conditions.

Case studies:

5) Line 15ff: you state that birch Pollen (20-30 μm) are smaller than spruce (70-100 μm) and therefore the Ångström exponent is higher when only birch is prevailing. However, considering the laser wavelength and the size of the particles, I would expect an Ångström exponent of 0 for all particles (size parameter well above 0). Can you state on this?

The Ångström exponent we are measuring is not the Ångström exponent of pure pollen. Also background aerosol contributes to the observed optical properties. We assume that pollen have a contribution to the Ångström, but the derivation from 0 is caused by the background aerosol. We are working on separation techniques of pollen and background aerosol to gain pure pollen properties without the effect of background aerosol, but this is not in the scope of this study.

6) Derived optical parameters: Are the Pollen probably aligned in a certain way that they can fly better and might therefore a difference in the observed optical properties in comparison to laboratory measurements occur?

Pollen grains with air bladder, e.g. spruce pollen, align with their air bladders upwards when drifting in the air. (Schwendemann, 2007) and also the almost spherical birch pollen were found to have an orientation when falling (Sassen, 2011; Tränke and Mielke, 1994). This could also partly explain differences between ambient measurements and laboratory experiments, this thought is added in the manuscript as:

Also the orientation of the pollen grains in the atmosphere has to be considered. Pollen with air bladders, e.g. spruce pollen, are known to align with their air bladders upwards when drifting in the air (Schwendemann et al., 2007) and also a certain orientation of the almost spherical birch pollen grains was observed (Sassen, 2011; Tränke and Mielke, 1994). This could cause differences in the measured optical properties if the orientation of

the particles in laboratory experiments is not considered and the irregularly shaped particles are observed from different angles.

Idea:

7) As you have determined the particle depolarization ratio of pure birch and you know the contribution of both constituents during the mixed birch/spruce period, couldn't you calculate the theoretical depolarization ratio of pure spruce by applying the depolarization separation formula of Tesche, 2009, JGR. If I am right, you will need to convert your number concentrations to surface concentrations to obtain the backscatter fraction.

This is a good idea, but we don't determine the particle depolarization of pure birch. We are providing the depolarization ratio for a period with birch pollen including background aerosol. The fraction/contribution of background aerosol cannot be determined with the setup during the presented campaign, which complicates the determination of the pure spruce depolarization. However, we are working on a method to estimate the pure PDR of different pollen and it will be the topic of a future paper which is going to be submitted soon.