Detection and characterization of birch pollen in the atmosphere using a multiwavelength Raman polarization lidar and Hirst-type pollen sampler in Finland

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Abstract. We present the results of birch pollen characterization using lidar and in situ measurements based on a 11 d pollination period from 5 to 15 May 2016 at the European Aerosol Research Lidar Network (EARLINET) station in Vehmasmäki (Kuopio; 62°44’ N, 27°33’ E), Finland. The ground-based multiwavelength Raman polarization lidar PollyXT performed continuous measurements at this rural forest site and has been combined with a Hirst-type volumetric air sampler, which measured the pollen type and concentration at roof level (4 m). The period was separated into two parts due to different atmospheric conditions and detected pollen types. During the first period, high concentrations of birch pollen were measured with a maximum 2 h average pollen concentration of 3700 grains m⁻³. Other pollen types represented less than 3 % of the total pollen count. In observed pollen layers, the mean particle depolarization ratio at 532 nm was 10 ± 6 % during the intense birch pollination period. Mean lidar ratios were found to be 45 ± 7 and 55 ± 16 sr at 355 and 532 nm, respectively. During the second period, birch pollen was still dominant, but a significant contribution of spruce pollen was observed as well. Spruce pollen grains are highly nonspherical, leading to a larger mean depolarization ratio of 26 ± 7 % for the birch–spruce pollen mixture. Furthermore, higher lidar ratios were observed during this period with mean values of 60 ± 3 and 62 ± 10 sr at 355 and 532 nm, respectively. The presented study shows the potential of the particle depolarization ratio to track pollen grains in the atmosphere.

1 Introduction

Atmospheric pollen is a well-known health threat as it can irritate the respiratory system and cause asthmatic symptoms (Bousquet et al., 2008). The number of people suffering from pollen-triggered diseases is rising (Schmidt, 2016), and the prevalence of pollen allergies is likely to further increase due to climate change as the pollination season becomes longer and pollen production increases (Lake et al., 2018). In addition to the well-known allergenic impacts, pollen also affects the climate (IPCC, 2013; WHO, 2003). Steiner et al. (2015) suggested that fragments of pollen act as cloud condensation nuclei (CCN) and therefore influence cloud optical properties. Pollen can furthermore change ice cloud formation processes by acting as ice nuclei (IN) (von Blohn et al., 2005; Diehl et al., 2001, 2002).

Worldwide, 879 active stations have continuously monitored pollen type and concentration near ground level in 2016 (Buters et al., 2018). The majority of these stations operate with Hirst-type volumetric air samplers. These traditional pollen traps are operated manually, which requires human resources and is time consuming. In recent years, novel techniques have been developed to enable automated pollen monitoring and reduce workload. Those techniques use, for example, automated image recognition (Oteros et al., 2015) or fluorescence spectra (Crouzy et al., 2016; Richardson et al., 2019; Saito et al., 2018) to identify pollen types, and they could enable a systematic pollen monitoring at ground level in near-real-time. Systematic information on the vertical dis-
Polarization Lidar Polly from the city center of Kuopio, Eastern Finland. The measurements took place from the beginning of May to the end of August 2016 at Vehmasmäki, Finland. Our measurement campaign took place from the beginning of May to the end of August 2016, the multiwavelength Raman polarization lidar PollyXT (Engelmann et al., 2010) performed continuous measurements at the rural forest station in Vehmasmäki (Kuopio), which is part of the European Aerosol Research Lidar Network (EARLINET). Simultaneously, a Hirst-type volumetric air sampler was operated to obtain pollen type and concentration at roof level. Twenty-one different pollen types were detected from May to August 2016. In this study, we focus on the description of birch pollen and the mixture of pollen types and the most allergenic tree pollen in northern, central and eastern Europe (D’Amato et al., 2010). Measurements were conducted with the multiwavelength Raman polarization lidar PollyXT (Althausen et al., 2009; Baars et al., 2016; Engelmann et al., 2016). PollyXT has three emission wavelengths (355, 532 and 1064 nm) and seven detection channels. In addition to the three emitted wavelengths, the backscattered signals at the inelastic Raman-shifted wavelengths (387, 407 and 607 nm) and the cross-polarized signal at 532 nm are detected. During nighttime, extinction and backscatter coefficient profiles at 355 and 532 nm can be determined independently using the Raman method (Ansmann et al., 1992). During daytime, the Klett–Fernald method (Fernald, 1984; Klett, 1981) is applied using the elastic signals due to the low signal-to-noise ratio for the Raman channels. The signal at the 407 nm Raman-shifted wavelength is used to determine water vapor mixing ratio profiles in air during mid-latitude conditions. The simultaneous measurement of the cross-polarized and total backscattered light at 532 nm enables the derivation of the linear particle depolarization ratio (PDR; Freudenthaler et al., 2009), which allows for the characterization of particle shape (Sassen, 2005). The measurement of multiple wavelengths allows for the retrieval of Ångström exponents (Å), which are related to the particle size. The ratio of extinction to backscatter coefficient is called lidar ratio (LR). It is considered an important criterion for particle characterization, as it depends on single-scattering albedo and backscatter phase function and therefore on the size distribution and the chemical composition of the aerosol particle. The LR is therefore considered to be aerosol-type dependent.

The operated lidar system has an initial spatial resolution of 30 m and a temporal resolution of 30 s. Due to the biaxial setup of emission and detection units, the height of complete overlap between the laser and the receiver field of view is reached at around 800–900 m (Engelmann et al., 2016). An overlap correction can be applied on the basis of a simple technique proposed by Wandinger and Ansmann (2002), which allows operators to extend profiles down to around 500 m. In this study, the lower limit of reliable profiles of vertically smoothed and temporally averaged optical properties is at around 800 m. Uncertainties in nighttime lidar products are mainly determined by signal noise and the correction of Rayleigh scattering. The overall relative errors of the lidar-derived optical properties retrieved with the Raman method are in the range of 5–10% for backscatter coefficients and depolarization ratios and 10–20% for extinction coefficients (Ansmann et al., 1992; Baars et al., 2012). These results are rare or missing. Light detection and ranging (lidar) is an effective method to investigate the vertical distribution of aerosols, as it enables measurements with high vertical and temporal resolutions under ambient conditions.
uncertainties propagate to the retrieved Ångström exponents and LRs.

Further details on the setup, principle and error propagation of Polly\textsuperscript{XT} can be found in Althausen et al. (2009) and Engelmann et al. (2016). Near-real-time measurements and Polly\textsuperscript{XT} data can be accessed at the PollyNET website (http://polly.tropos.de/, last access: 22 November 2019).

2.2 Pollen collector: Hirst-type volumetric air sampler

A Hirst-type volumetric air sampler located next to the lidar, 4 m above ground, monitored the pollen concentration and type. This type of spore sampler enables continuous 7 d collection of pollen grains with 2 h time resolution. The sampling principle is based on the design described by Hirst (1952). With a flow rate of 10 L min\textsuperscript{−1}, air is drawn into the sampling device through a 14 mm × 2 mm orifice. A large wind vane on a rotatable sampler head makes the sampler sensitive to changes in wind direction and ensures that the orifice is always oriented towards the wind. Particles impact on an adhesive-coated plastic tape beneath the orifice. For this study, the tape, fixed on a rotating drum, was changed every 7 d and the pollen grains impacted on the tape were further analyzed under the microscope. The pollen type was determined using characteristic features of the examined pollen grains. By converting the counted spores on the sample tape surface in relation to the inlet air flow, the pollen concentration was obtained.

3 Methodology

Figure 1 shows the temporal variation in the pollen concentration (Fig. 1a), the range-corrected signal at 1064 nm (Fig. 1b) and the volume depolarization ratio at 532 nm (Fig. 1c) during the period 5–15 May 2016. This period represents the main birch pollination season of 2016 as 83 % of the annual birch pollen had been collected during that time. A relatively large aerosol load was observed within the planetary boundary layer up to ~ 3.5 km. As shown in Fig. 1c, the volume depolarization ratio ranges between 4 % and 10 % suggesting the presence of nonspherical particles. A detailed examination of the air masses arriving during this period along with modeled dust load using the BSC-DREAM8b model (Basart et al., 2012) confirms the absence of dust in middle and northern Europe. Additionally, MODIS data (MODIS, 2019) were synergistically used to exclude smoke aerosol layers from biomass burning. Hence, the highly depolarizing aerosol layers were likely attributed to pollen, keeping in mind that some contamination with local anthropogenic aerosol is always possible.

Ground-level pollen concentration values presented in Fig. 1a were used to verify the strong pollination event in the beginning of May, which provides 50 % of the annual birch pollen concentration. The event started in the evening (17:00 UTC) of 5 May and lasted until noon on 9 May (hereafter called period 1). During period 1, the 2 h average pollen concentration exceeded 1000 grains m\textsuperscript{−3} for 53 % of the time. The majority of pollen identified was birch (97 %) with a very small contribution from willow (2 %) and other pollen types (1 %). From 12 to 15 May (period 2), the mean pollen concentration was significantly lower. Only 8 % of the time, the total pollen concentration was higher than 1000 grains m\textsuperscript{−3}. In addition to birch (82 %), spruce pollen (14 %) and other pollen types (4 %) were detected. This variation can be explained by the different meteorological conditions during the two periods. A different predominant wind direction during the two periods was observed, which probably caused the different mixtures of pollen types. The most frequent wind direction in period 1 was northwest, whereas in period 2 the air masses were mainly advected from the southeast. When comparing the diurnal cycle of temperature and relative humidity measured at ground level, we found higher temperature values and lower relative humidity during period 1 compared with period 2. Temperature and pollen concentration have been shown to be positively correlated, whereas pollen concentration and relative humidity show a negative correlation (Bartková-Števková, 2003). The different pollen concentrations could therefore be partly explained by variations in temperature and humidity.

The near-ground aerosol layers are assumed to contain the highest concentration of local pollen, and they are defined as pollen layers in this study. The gradient method was applied to determine the bottom and top layer heights of the pollen layers (Bösenberg and Matthias, 2003; Flamant et al., 1997; Mattis et al., 2008). The local maximum in the first derivative of the 1064 nm backscatter coefficient was considered to be the bottom of the layer. The local minimum was considered to be the layer top. To verify the determined layers, the layer boundaries identified by the gradient method were compared with the bottom and top heights of coherent structures in the height–time illustration of the range-corrected signal (Giannakaki et al., 2015). The layer identification was based on the assumption that the optical properties should be relatively homogeneous, which means that within one layer the variability of the optical properties should be lower than the statistical uncertainty of the individual data points. Two layers with a vertical distance less than 100 m apart from each other were combined into one layer. All layers detected during the 11 d period are shown in Fig. 2. Black, magenta, blue and yellow bars show the first, second, third and fourth layers, respectively. Triangles mark the part of the layer which was used for calculations of the mean optical properties of the layer. The lower limit for reliable profiles during our measurement period was at around 800 m. Since the closest layer to the ground is assumed to contain the highest pollen concentration and share, we only consider the lowest layers (black) in the following analysis.
4 Results and discussion

4.1 Case studies

We present two case studies representative for different pollen mixtures: in the first case study only birch pollen had been detected by the Hirst-type sampler, and in the second case study spruce pollen was detected in addition to birch. In the choice of case studies, backward trajectories have been considered to select cases with minimal contamination with other aerosol. Furthermore, nighttime Raman measurements were chosen to present all lidar-derived parameters including the retrieved LR profile. Figure 3 shows, from left to right, the particle backscatter coefficient at 355 (blue), 532 (green) and 1064 nm (red); the particle extinction coefficient at 355 (dashed blue) and 532 nm (dashed green); the LR at 355 (blue) and 532 nm (green); the PDR at 532 nm (light green); the Ångström exponents calculated both from the backscatter coefficient at 355–532 nm (blue) and 532–1064 nm (red) and from extinction coefficients at 355–532 nm (black); and the relative humidity from lidar-derived water vapor profiles (black) and temperature profiles from a radiosonde launched at 18:00 UTC (orange). Lidar-derived optical properties were vertically smoothed using a sliding average of 25 bins (750 m). Four-day backward trajectories ending at the height of the layers and the middle of the time period are shown as well.
Figure 3. Two case studies of different pollen mixtures. (a, b) Period 1, 6 May 2016 23:00–01:00 UTC, only birch pollen was collected by the Hirst-type volumetric air sampler. (c, d) Period 2, 15 May 2016 19:00–21:00 UTC, birch and spruce pollen were collected. Profiles of backscatter and extinction coefficients, particle depolarization and lidar ratio, Ångström exponents, relative humidity (derived from lidar measurements) and temperature profiles (18:00 UTC radio sounding). (b, d) Four-day HYSPLIT backward trajectories. Defined pollen layers are marked in gray.

The first case study was selected during the intense birch pollination event (period 1). On 6 May 2016 between 23:00 and 01:00 UTC only birch pollen was detected. Using the layer definition methodology (Sect. 3), three layers were determined, and the two lowest ones have been combined to one pollen layer for this analysis since the distance was less than 100 m.

Four-day HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) backward trajectories (Stein et al., 2015) ending at 450 m and 1.1 km on 7 May 00:00 UTC show that the air masses are advected from western directions and have traveled over the British Isles, the North Sea and southern Sweden. The contamination with depolarizing aerosol like dust is therefore considered to be negligible; however, the mixture with other anthropogenic aerosol cannot be ruled out. The presumed birch pollen layer was observed up to 1.2 km. Extinction coefficient at 355 nm is about 22 ± 2 and is 13 ± 1 Mm⁻¹ at 532 nm. The mean LR for the observed layer is 49 ± 4 and 70 ± 7 sr at 355 and 532 nm, respectively. Mean backscatter and extinction-related Ångström exponents at 355–532 nm are 2.1 ± 0.04 and 1.1 ± 0.5, respectively. The backscatter-related Ångström exponent between 532 and 1064 nm is 0.9 ± 0.1. The mean PDR at 532 nm within the layer is 14 ± 1 %. Note that it can be even higher close to the ground, below the height of complete overlap. The PDR decreases with increasing height, while the LR remains constant. Thus the measured LR may not be a good indicator for characterizing the observed birch pollen in these cases as the contribution of pollen is assumed to decrease with increasing distance to the pollen source.

During our second case study on 15 May 2016 between 19:00 and 21:00 UTC, spruce pollen had been measured simultaneously with the birch pollen. Profiles and backward trajectories are shown in the lower row of Fig. 3 (Fig. 3c and d). The pollen layer reaches up to 1.7 km.

The extinction coefficients at 355 and 532 nm are higher than in the previous case, being 61 ± 5 Mm⁻¹ at 355 nm and 44 ± 6 Mm⁻¹ at 532 nm. The mean LR is 55 ± 6 sr at 355 nm and 51 ± 9 sr at 532 nm. The backscatter and extinction-related Ångström exponents at 355–532 nm are lower than in the first case with values of 0.5 to 0.7 and 0.1 to 1.7,
and analyzed to determine the relationship between pollen layers and the long-distance transport of birch pollen. All pollen layers between 5 and 15 May have been identified and analyzed to determine the relationship between pollen layers and the long-distance transport of birch pollen. All pollen layers between 5 and 15 May have been identified and analyzed to determine the relationship between pollen layers and the long-distance transport of birch pollen. All pollen layers between 5 and 15 May have been identified and analyzed to determine the relationship between pollen layers and the long-distance transport of birch pollen. All pollen layers between 5 and 15 May have been identified and analyzed to determine the relationship between pollen layers and the long-distance transport of birch pollen. All pollen layers between 5 and 15 May have been identified and analyzed to determine the relationship between pollen layers and the long-distance transport of birch pollen. 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be used as an indicator for detecting the presence of pollen. The Ångström exponent, on the other hand, can be also related to the amount and type of background aerosol and is therefore less representative here.

Earlier studies show that relative humidity can affect the size and shape of pollen grains, which leads to different optical properties (Franchi et al., 1984; Griffiths et al., 2012; Katifori et al., 2010). When pollen grains dehydrate, the pollen wall can fold onto itself to prevent further dehydration, and this phenomena is known as harmomegathy (Katifori et al., 2010). The shape of the pollen grain changes, which could lead to significantly higher depolarization of the backscattered light. At humid conditions, pollen grains swell by taking up water internally and after reaching a relative humidity over 89 % external wetting of the pollen surface can occur (Griffiths et al., 2012). To check whether the ambient relative humidity affects our measurements, the Ångström exponent (532–1064 nm) and the PDR at 532 nm are presented against the relative humidity in Fig. 6. In the selected measurement periods, the relative humidity ranged between 40 % and 65 %. In this humidity range, Ångström exponent (Fig. 6a) and depolarization ratio (Fig. 6b) do not show any correlation with the relative humidity. Thus, our measurements were not affected by extreme humidity events and represent values for pollen under ambient atmospheric conditions in the spring season in Finland. However, lidar measurements of relative humidity profiles are only available during nighttime. The relative humidity in the observed pollen layers during daytime could be smaller. This could result in occasional folding of the pollen grains and higher depolarization ratios. This hypothesis could also explain the higher depolarization ratios of about 25 % of a few Klett measurements of birch pollen during the first period.

Table 1 summarizes the mean intensive properties together with the associated standard derivation (SD), range and median in the first (birch) period of our campaign. The contribution of other pollen types in this period was small. Those values, therefore, can be considered to be characteristic for birch-pollen-dominated aerosol conditions. Table 2 shows the same properties for the spruce-contaminated period. Lidar ratio and PDR are higher when spruce is detected simultaneously with birch. The PDR values for birch pollen are considerably lower than previously determined in lidar studies. A linear depolarization ratio up to 30 % at 694 nm was detected by Sassen (2008) for paper birch in Alaska. And under controlled laboratory environment, Cao et al. (2010) measured a linear depolarization ratio at 532 nm of 33 % for dried paper birch pollen. We assume that those high depolarization values can be caused by dry birch pollen grains, which fold and change their shape when dehydrating. Under ambient conditions the pollen grains are more spherical and therefore less depolarizing. Also, the orientation of the pollen grains in the atmosphere has to be considered. Pollen with air bladders, e.g., spruce pollen, is known to align with its air bladders upwards when drifting in the air (Schwendemann...
Table 1. Mean values, range and median of optical properties of the detected pollen layers in the first measurement period: 5–10 May, intense birch pollination period.

| Parameter                  | Mean ± SD | Range     | Median |
|----------------------------|-----------|-----------|--------|
| Layer top height (km)      | 1.3 ± 0.3 | 1.0–2.2   | 1.2    |
| Backscatter coefficient    |           |           |        |
| (Mm\(^{-1}\)sr\(^{-1}\)) | 355 nm    | 0.7 ± 0.5 | 0.1–2.4 | 0.7    |
|                            | 532 nm    | 0.3 ± 0.2 | 0.1–1.0 | 0.3    |
|                            | 1064 nm   | 0.2 ± 0.1 | 0.1–0.4 | 0.2    |
| Extinction coefficient     |           |           |        |
| (Mm\(^{-1}\))             | 355 nm    | 33.0 ± 13.3 | 20.0–68.2 | 30.9  |
|                            | 532 nm    | 19.0 ± 6.5 | 11.0–34.4 | 19.1  |
| Lidar ratio (sr)           |           |           |        |
|                            | 355 nm    | 46 ± 8    | 34–60   | 46     |
|                            | 532 nm    | 52 ± 12   | 31–74   | 53     |
| PDR                        |           |           |        |
|                            | 532 nm    | 0.10 ± 0.06 | 0.03–0.26 | 0.08  |
| Number of pollen layers    |           |           |        |
| all: 41                    | Raman: 10 |

Table 2. Mean values, range and median of optical properties of the detected pollen layers in the second measurement period: 12–15 May, spruce-contaminated period.

| Parameter                  | Mean ± SD | Range     | Median |
|----------------------------|-----------|-----------|--------|
| Layer top height (km)      | 1.3 ± 0.4 | 1–2.2     | 1.1    |
| Backscatter coefficient    |           |           |        |
| (Mm\(^{-1}\)sr\(^{-1}\)) | 355 nm    | 0.7 ± 0.2 | 0.3–1.1 | 0.6    |
|                            | 532 nm    | 0.5 ± 0.2 | 0.3–0.8 | 0.4    |
|                            | 1064 nm   | 0.3 ± 0.1 | 0.2–0.4 | 0.2    |
| Extinction coefficient     |           |           |        |
| (Mm\(^{-1}\))             | 355 nm    | 52.9 ± 13.1 | 26.9–60.9 | 58.5  |
|                            | 532 nm    | 40.0 ± 9.5 | 24.6–54.6 | 40.2  |
| Lidar ratio (sr)           |           |           |        |
|                            | 355 nm    | 60 ± 3    | 55–64   | 59     |
|                            | 532 nm    | 62 ± 10   | 49–77   | 60     |
| PDR                        |           |           |        |
|                            | 532 nm    | 0.26 ± 0.07 | 0.18–0.39 | 0.24  |
| Number of pollen layers    |           |           |        |
| all: 12                    | Raman: 5  |

et al., 2007), and also an orientation of the almost spherical birch pollen grains was observed (Sassen, 2011; Tränkle and Mielke, 1994). This could cause differences in the measured optical properties if the orientation of the particles in laboratory experiments is not considered, or if the irregularly shaped particles are observed from different angles.

5 Conclusion

Particle depolarization ratios of about 10 % have been observed during a birch pollination event in Vehmasmäki, Finland. When more nonspherical pollen, e.g., spruce, is present, the particle depolarization ratio can be as high as 26 %. Those depolarization ratios are similar to dust and biomass-burning aerosol mixtures (Tesche et al., 2011) or dust mixtures with marine aerosol (Groß et al., 2011), thus pollen could easily be misclassified as dusty mixtures. The mean LRs show a wide range of values depending on the mixing of different pollen types in the atmosphere. The mean LR at 355 nm varies between 46 ± 8 sr (first period) and 60 ± 3 sr (second period) and at 532 nm between 52 ± 12 sr (first period) and 62 ± 10 sr (second period). Those LRs are characteristic for dust or dust–smoke mixtures (Tesche et al., 2011), which complicates the characterization of pollen using the LR. Also the backscatter-related Ångström exponents at 532–1064 nm, which is around 1.0 for the intense birch pollination period and around 0.8 for the spruce-contaminated period, are similar to characteristic values for smoke and dust–smoke mixtures, respectively. Thus, in order to distinguish between pollen and other aerosol types, all three parameters and backward trajectories as well as possible dust and biomass-burning aerosol sources have to be considered.

The presented data show the potential of lidar measurements to detect pollen in the atmosphere. Nevertheless, there are challenges which need to be addressed in order to improve the characterization of optical properties of airborne
pollen. First, the minimum height of the usable lidar signal needs to be as low as possible. By operating a lidar system with a low full-overlap height or additional near-field channels, the coverage of lower heights can be significantly improved. Second, the contribution of other aerosol types like anthropogenic pollution has to be determined. Therefore, more multiwavelength lidar studies with depolarization characterization on atmospheric pollen are necessary.

Data availability. Lidar data are available upon request from the authors and data “quicklooks” are available on the PollyNET website (http://polly.tropos.de/, last access: 22 November 2019). Trajectories are calculated with the NOAA (National Oceanic and Atmospheric Administration) HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model (https://ready.arl.noaa.gov/HYSPLIT.php, last access: 30 April 2019). Fire data are available at the NASA Worldview application (https://worldview.earthdata.nasa.gov, last access: 30 April 2019). BSC-DREAM8b model simulations are operated by the Barcelona Supercomputing Center and are available at https://ess.bsc.es/bsc-dust-daily-forecast/ (last access: 30 April 2019).

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