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CONTRIBUTIONS

CHARACTERIZATION OF ATMOSPHERIC POLLEN WITH ACTIVE REMOTE SENSING

STEPHANIE BOHLMANN

FINNISH METEOROLOGICAL INSTITUTE
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No. 175

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Stephanie Bohlmann

Finnish Meteorological Institute
Atmospheric Research Centre of Eastern Finland
Kuopio, Finland

ACADEMIC DISSERTATION

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Abstract

Atmospheric pollen is a well-known health threat causing allergy-related diseases. As a biogenic aerosol, pollen also affects the climate by directly absorbing and scattering solar radiation and by acting as cloud condensation or ice nuclei. A good understanding of pollen distribution and transport mechanisms is needed to evaluate the environmental and health impacts of pollen. However, pollen observations are usually performed close to ground and vertical information, which could be used to evaluate and improve pollen transport models, is widely missing.

In this thesis, the applicability of lidar measurements to detect pollen in the atmosphere is investigated. For this purpose, measurements of the multiwavelength Raman polarization lidar PollyXT at the rural forest site in Vehmasmäki (Kuopio), Eastern Finland have been utilized. The depolarization ratio was identified to be the most valuable optical property for the detection of atmospheric pollen, as non-spherical pollen like pine and spruce pollen causes high depolarization ratios. However, detected depolarization ratios coincide with typical values for dusty mixtures and additional information such as backward trajectories need to be considered to ensure the absence of other depolarizing aerosols like dust. To separate pollen from background aerosol, a method to estimate the optical properties of pure pollen using lidar measurements was developed. Under the assumption that the Ångström exponent of pure pollen is zero, the depolarization ratio of pure pollen can be estimated. Depolarization ratios for birch and pine pollen at 355 and 532 nm were determined and suggested a wavelength dependence of the depolarization ratio. To further investigate this wavelength dependence, the possibility to use depolarization measurements of Halo Doppler lidars (1565 nm) was explored. In the lower troposphere, Halo Doppler lidars can provide reasonable depolarization values with comparable quality to PollyXT measurements. Finally, measurements of PollyXT and a Halo StreamLine Doppler lidar were used to determine the depolarization ratio at three wavelengths. A wavelength dependence of the particle depolarization ratio with maximum depolarization at 532 nm was found. This could be a characteristic feature of non-spherical pollen and the key to distinguish pollen from other depolarizing aerosol types.

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Nimeke
Ilmakehän siitepölyjen havainnointi kaukokartoitusmenetelmin

Tiivistelmä

Ilmakehän siitepölyt vaikuttavat sekä ihmisten terveyteen allergisten sairauksien kautta, että biogeenisenä aerosolina ilmastoon absorpoimalla sekä sirottamalla auringon säteilyä ja toimimalla tiivistymisytiminä pilville tai jääkiteille. Jotta siitepölyjen terveys- ja ympäristövaikutuksia voitaisiin ymmärtää paremmin, tarvitaan tietoa siitepölyn jakautumisesta sekä kulkeutumismekanismeista. Yleensä siitepölyjä havainnoidaan kuitenkin vain maan tasalla, eikä siitepölyjen pystysuuntaisesta jakautumisesta ole mitattua tietoa. Tätä tietoa voitaisiin kuitenkin hyödyntää siitepölyjen kulkeutumismallien verifiointissa sekä kehittämisessä.

Tässä työssä tarkastellaan lidar-mittausten käytettävyyttä siitepölyn havaitsemiseen ilmakehässä. Tätä varten hyödynnettiin usean aallonpituuden Raman-polarisaatio PollyXT lidaria, joka sijaitsee Vehmasmäen mittausasemalla Kuopiossa. Depolarisaation huomattiin olevan hyödyllisin optinen ominaisuus siitepölyjen tunnistamiseen, sillä ei-pallomaiset siitepölyhiukkaset aiheuttavat suuren depolarisaatio-signaalin. Havaitut depolarisaatioarvot ovat kuitenkin tyypillisiä myös aavikkopölylle, joten näiden erottamiseksi tulee tarkastella esimerkiksi mitatun ilmamassan kulkeutumista jotta voidaan poissulkea muiden vahvasti polarisoivien aerosolien esiintyminen. Siitepölyn erottamiseksi ilmakehän muista tausta-aerosoleista kehitettiin menetelmä jolla voidaan arvioida pelkän siitepölyn optisia ominaisuuksia lidar-mittauksista. Oletettaessa Ångströmin eksponentin olevan nolla siitepölylle, voidaan siitepölyn aiheuttama depolarisaatio arvioida. Menetelmällä arvioitiin depolarisaatio-arvot männyn ja kuusen siitepölylle 355 nm:n ja 532 nm:n aallonpituuksilla ja havaittiin selkeä aallonpituusriippuvuus depolarisaation suhteen. Tämän riippuvuussuhteen syvempää tarkastelua varten selvitettiin mahdollisuuksia Halo Doppler lidareiden depolarisaatio-mittausten (1565 nm aallonpituus) käyttöön. Alemmassa troposfäärissä Halo Doppler lidarit voivat tuottaa luotettavia depolarisaatio-mittauksia verraten PollyXT-mittauksiin. Viimeisessä vaiheessa hyödynnettiin depolarisaatio-mittauksia PollyXT:stä ja Halo StreamLine Doppler lidarista kolmella aallonpituudella. Tutkimuksessa havaittiin sekä depolarisaation aallonpituusriippuvuus että suurin depolarisaatio 532 nm:n aallonpituudella. Tämä voi olla ei-pallomaisen siitepölyjen tunnistettava ominaisuus ja avain siitepölyjen erottamiseen muista vahvasti polarisoivista aerosoleista.

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List of Acronyms

AD-NET	Asian Dust Network
APS	Aerodynamic particle sizer
CALIOP	Cloud-Aerosol Lidar with Orthogonal Polarization
CALIPSO	Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations
CCN	Cloud condensation nuclei
DIAL	Differential absorption lidars
EARLINET	European Aerosol Research Lidar Network
EarthCARE	Earth Clouds, Aerosol and Radiation Explorer
FAO	Food and Agriculture Organization
FMI	Finnish Meteorological Institute
FOV	Field-of-view
GDAS	Global Data Assimilation System
INP	Ice nucleating particles
IPCC	Intergovernmental Panel on Climate Change
IPP	Intense pollination period
LALINET	Latin America Lidar Network
LIDAR	Light Detection And Ranging
LIF	Laser induced fluorescence
LR	Lidar ratio
MPLNET	Micro-Pulse Lidar Network
ND:YAG	Neodymium-doped yttrium aluminum garnet
NDACC	Network for the Detection of Atmospheric Com- position Change
OPS	Optical particle sizer
PDR	Particle depolarization ratio
PMT	Photomultiplier tube
PollyXT	PORtabLe Lidar sYstem with eXTended capabilities
Rapid-E	Real-Time Airborne Particle Identifier
SHARP	Synchronized Hybrid Ambient Real-time Particu- late Monitor
SHG	Second harmonic oscillator
SMPS	Scanning mobility particle sizer
SNR	Signal-to-noise ratio
THG	Third harmonic oscillator
WAO	World Allergy Organization
WHO	World Health Organization
WIBS	Wideband Integrated Bioaerosol Sensor

List of original publications

- I Bohlmann, S., X. Shang, E. Giannakaki, M. Filioglou, A. Saarto, S. Romakkaniemi, and M. Komppula (2019), Detection and characterization of birch pollen in the atmosphere using a multiwavelength Raman polarization lidar and Hirst-type pollen sampler in Finland, *Atmospheric Chemistry and Physics*, 19(23), 14559–14569, doi: 10.5194/acp-19-14559-2019
- II Shang, X., E. Giannakaki, S. Bohlmann, M. Filioglou, A. Saarto, A. Ruuskanen, A. Leskinen, S. Romakkaniemi, and M. Komppula (2020), Optical characterization of pure pollen types using a multi-wavelength Raman polarization lidar, *Atmospheric Chemistry and Physics*, 20(23), 15323–15339, doi: 10.5194/acp-20-15323-2020
- III Vakkari, V., H. Baars, S. Bohlmann, J. Bühl, M. Komppula, R.-E. Mamouri, and E. J. O'Connor (2021), Aerosol particle depolarization ratio at 1565 nm measured with a Halo Doppler lidar, *Atmospheric Chemistry and Physics*, 21(8), 5807–5820, doi: 10.5194/acp-21-5807-2021
- IV Bohlmann, S., X. Shang, V. Vakkari, E. Giannakaki, A. Leskinen, K. E. J. Lehtinen, S. Pätsi, and M. Komppula (2021), Lidar depolarization ratio of atmospheric pollen at multiple wavelengths, *Atmospheric Chemistry and Physics*, 21(9), 7083–7097, doi: 10.5194/acp-21-7083-2021

1 Introduction

The Earth's atmosphere, the layer of gases surrounding our planet, does not only protect us from the hazards of space and regulate the surface temperature to a habitable level, it is also the stage for a multitude of processes that affect our everyday life. The atmosphere is composed of a mixture of gases, among which nitrogen and oxygen are the most abundant ones followed by argon, carbon dioxide, water vapor and other trace gases. Within this gas mixture, small solid or liquid particles are suspended. This suspension of solid or liquid particles in air is called aerosol. Aerosols are omnipresent in the atmosphere, but their concentration is extremely variable due to the diversity of aerosol sources and the relatively short residence time in the atmosphere which ranges from hours to weeks (Boucher, 2015). Aerosol sources can be of natural or anthropogenic origin. Natural aerosols are for example mineral (desert) dust, volcanic ash, sea spray, biomass burning aerosol or biogenic aerosols. Anthropogenic aerosols, on the other hand, are mainly emitted by the combustion of fossil fuels or biofuels.

In the atmosphere, aerosols significantly affect atmospheric processes and influence the weather and climate on Earth. By scattering and absorbing solar and terrestrial radiation, aerosols directly affect the radiation budget (Haywood and Boucher, 2000). The absorption of radiation by atmospheric aerosols affects the temperature profile in the atmosphere and therefore the relative humidity and atmospheric stability which thereby affects the formation and lifetime of clouds. This effect is known as semi-direct aerosol effect (Hansen et al., 1997; Johnson et al., 2004). In addition to direct and semi-direct aerosol radiative forcing, aerosols also influence the climate indirectly by acting as cloud condensation nuclei (CCN) or ice nucleating particles (INP) and consequently modifying microphysical and optical properties (Twomey, 1977) and the lifetime of clouds (Albrecht, 1989). The effects of aerosols on the climate are difficult to quantify as processes partially counteract each other and are intertwined with meteorological variability, thereby the uncertainties in aerosol–cloud interactions and the related radiative forcing are large (IPCC, 2013). Since the first IPCC assessment report in 1990, the understanding of aerosol effects on the climate has steadily increased. More interaction modes and aerosol types have been investigated, but the estimates of aerosol radiative forcing are still uncertain (Bender, 2020).

Besides the impact on climate, aerosols also affect our life and health directly. Large concentrations of aerosols can significantly decrease visibility and restrict daily activities. Poor air quality caused by high amounts of particulate matter can lead to a variety of health effects, for example asthma, heart diseases, strokes or pregnancy complications (Bové et al., 2019; Kelly and Fussell, 2015). Also biogenic aerosols like pollen can cause mild to serious health problems and com-

promise life quality. Sneezing, coughing, reddened eyes, an itchy nose or throat or even shortness of breath and chest tightness are symptoms of allergic rhinitis, also known as hay fever. Already in the beginning of the 19th century these symptoms have been described by Bostock (1819). In the 1870s, pollen has then been identified as the cause of these symptoms by Blackley (1873). Since then, major effort has been done to characterize allergenic pollen, identify and describe pollen types and transport mechanisms and to predict pollen seasons and distribution patterns. Today airborne pollen is recognized as one of the major causes of allergy-related diseases such as asthma, rhinitis, eczema and conjunctivitis (Bousquet et al., 2008). According to the World Allergy Organization (WAO) about 10–30% of the global adult population and 40% of children are affected by allergic rhinitis and around 300 million people suffer from asthma (Pawankar et al., 2013). The number of people suffering from pollen allergies is expected to rise (Schmidt, 2016), not least because of the changing climate and expected longer growing seasons.

Pollen carries the male genetic material of flowering and seed-producing plants and is transported by animals (zoophilous), wind (anemophilous) or water (hydrophilous) from the stamens to the pistil or from the male to female cone. Anemophilous pollen poses a major risk in terms of pollen allergies as it is emitted in large quantities and can travel over long distances (Bousquet et al., 2008).

Dispersed in the air anemophilous pollen does not only find its way into the respiratory system of humans, it also contributes to the aerosol load in the atmosphere. Atmospheric pollen as one kind of biogenic aerosol has been the focus of several studies. Pope (2010) and Steiner et al. (2015) investigated the ability of pollen to act as CCN. Pollen as INP for the different freezing modes has been investigated by Diehl et al. (2001, 2002) and von Blohn et al. (2005). These studies found that pollen and its fragments can act as CCN and INP and can therefore influence cloud formation and subsequently the climate. Although the global contribution of pollen to cloud processes is assumed to be low, pollen could be of importance on regional and seasonal scale (Hoose et al., 2010). To investigate and improve the understanding of the distribution of atmospheric pollen is therefore not only valuable for allergy sufferers but also for the scientific community.

The distribution of pollen is usually monitored on roof-level (i.e. close to the ground) with so-called Hirst-type pollen samplers, though other innovative detection systems, for example using automatic image recognition, have been developed in recent years. Nonetheless, pollen detection systems are usually bound to ground and vertical pollen information is only sporadically obtained by airborne campaigns (Damialis et al., 2017; Mandrioli et al., 1984; Rempe, 1937). However, the vertical distribution is needed to validate and improve pollen transport and

forecast models.

One method predestined for the continuous and real-time profiling of aerosols in the atmosphere is Light Detection And Ranging (lidar). Lidars are active remote-sensing instruments that use the atmospheric scattering and extinction of light to attain information of the atmosphere's composition and state. Light can be emitted and detected at different wavelengths and polarization states which allows the derivation of optical properties of the backscattering object with high vertical and temporal resolution under ambient conditions. Measured properties are therefore not affected by the measurement itself. Different lidar systems have been developed to cover different regions of the atmosphere and tackle different research questions. Elastic and inelastic (Raman) lidars are used to detect the presence of aerosols and clouds and can provide vertical information of aerosol layers in the atmosphere. More advanced Raman lidars can also measure temperature and atmospheric gases, such as water vapor (Behrendt, 2005; Wandinger, 2005b). Differential absorption lidars (DIAL) are used for the detection of atmospheric gases and temperature profiling (Bösenberg, 2005; Gimmestad, 2005). Resonance fluorescence lidars can measure atoms and ions in the mesosphere (Abo, 2005) and Doppler lidars are used to measure turbulence and wind in the lower atmosphere (Werner, 2005).

Networks of ground-based lidar stations have been established all over the world to create a database of the vertical distribution of atmospheric components. One of them is the European Aerosol Research Lidar Network (EARLINET) (Pappalardo et al., 2014). Established in 2000, EARLINET includes currently 31 stations all over Europe which provide regular lidar measurements. Other lidar networks are for example the Asian Dust Network (AD-NET) (Sugimoto et al., 2016) with around 20 stations in East Asia and the Latin America Lidar Network (LALINET) (Antuña-Marrero et al., 2017) which comprises around 10 stations in South America. Global operating networks are, for example, the NASA Micro-Pulse Lidar Network (MPLNET) (Welton et al., 2001) and the Network for the Detection of Atmospheric Composition Change (NDACC) (Leblanc et al., 2016). Besides those ground-based lidar stations organized in lidar networks, lidars are also operated on-board aircraft (airborne) and on-board of satellites (spaceborne). The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) flying on-board the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite provides vertical profiles of aerosols on a global scale (Winker et al., 2003). And also the planned EarthCARE (Earth Clouds, Aerosols and Radiation Explorer) satellite is going to provide more valuable information of the vertical contribution of aerosols using a 355 nm spaceborne lidar (Illingworth et al., 2015).

Automatic aerosol classification schemes for spaceborne lidar observations (Groß et al., 2015; Kim et al., 2018), but also for ground-based lidar networks (Baars et al., 2017; Nicolae et al., 2018) enable the characterization of the aerosols distribution in near real-time. This information is necessary to determine the impact of specific aerosols on the climate and evaluate climate models. However, biogenic aerosols are not included in current automatic aerosol classification algorithms for ground based or spaceborne lidars and pollen is likely misclassified as dusty mixtures. Including airborne pollen in automatic classification schemes would not only improve the correct classification of aerosol types, it would also improve the understanding of pollen transport and distribution, as at the moment vertical information about pollen concentration is limited to occasional airborne or lidar campaigns. Spaceborne lidars and existing lidar networks could be used to detect pollen in the vertical with high spatial coverage. However, a better understanding of pollen detection with lidar is needed before pollen can be included in aerosol classification algorithms.

Few studies have explored the detection of pollen with lidar so far. In 2008, Sassen (2008) detected birch pollen in Alaska using a 694 nm polarization lidar. Noh et al. (2013a,b) found signatures of pollen in the atmosphere over South Korea using a 532 nm lidar and Sicard et al. (2016) investigated the vertical distribution of pollen over Barcelona. Using laser induced fluorescence (LIF) lidars, Saito et al. (2018) and Richardson et al. (2019) were able to detect the fluorescence spectrum of pollen in the atmosphere. Long-term measurements and observations at multiple wavelengths however are missing.

In this thesis, we explore the capabilities of multiwavelength Raman polarization lidar measurements to detect and characterize atmospheric pollen (**Paper I**). We present an approach to estimate the optical properties of pure pollen (**Paper II**). Furthermore, the possibility to use depolarization measurements of Doppler lidar instruments for pollen detection is shown (**Paper III**) and the use of depolarization ratio measurements at multiple wavelengths for pollen detection is presented (**Paper IV**). The articles contribute to the following research questions:

- 1) Can we detect and characterize atmospheric pollen using multiwavelength Raman polarization lidar? Which particle optical properties are most suitable for the detection of pollen?
- 2) Can we use lidar measurements to derive the depolarization ratio of pure pollen in the atmosphere?
- 3) Can we utilize Halo Doppler lidar measurements for the detection of pollen?

- 4) Can the detection of the lidar depolarization ratio at multiple wavelengths improve the characterization of atmospheric pollen? Can pollen types be separated using lidar optical properties at multiple wavelengths?

The thesis is organized as follows: An introduction to atmospheric pollen and the observations at our measurement site is given in Chapter 2. The theoretical background of the lidar technique with focus on the retrieval of the optical quantities is given in Chapter 3. The main results of the original publications are summarized in Chapter 4 and concluding remarks are given in Chapter 5.

2 Atmospheric pollen and its observation

Pollen is the part of angiosperms and gymnosperms that carries the male genetic material of the plant. It is produced in the male cone of gymnosperms or in the anthers of an angiosperm flower. Because plants themselves are immobile, the genetic material has to be transported by other means. The most common way of pollination is the transfer by animals (zoophily), especially by honey bees and other insects. But pollen can also be transported by wind (anemophily) or water (hydrophily) (Pacini, 2008). Most grasses (Poaceae), conifers and around 10-20% of flowering plants are anemophilous (Ackerman, 2000). Hydrophilous pollination is much less common than zoophily and anemophily and has been observed, for example, for seagrasses.

Pollen grains appear in a wide range of sizes, shapes and surface textures, depending on different pollination requirements and ways of transport. The variety of sizes spans almost over three orders of magnitude. The smallest pollen grains, the pollens of forget-me-not (*Myosotis spp.*), are only 5 μm in diameter (Leuschner, 1993), while pollen grains of *Oenothera* species have a diameter of around 167 μm (Knight et al., 2010). Generally, the pollen size, defined as the largest diameter, is categorized in very small ($<10 \mu\text{m}$), small (10-25 μm), medium-sized (26-50 μm), large (51-100 μm) and very large ($>100 \mu\text{m}$) (Halbritter et al., 2018). It has been noted that even small pollen grains are considered coarse particles in aerosol research, where particles with a diameter less than 1 μm are usually considered fine mode and above 1 μm coarse mode particles. Pollen is therefore rather big compared to other common aerosols e.g. mineral dust (100 nm-10 μm) or biomass burning aerosol ($<1 \mu\text{m}$) (Boucher, 2015). However, pollen grains can also rupture to smaller, fine mode, fragments under certain meteorological conditions, e.g. humidity changes and turbulent disturbances. These fragments, though, are usually not recorded as pollen using commonly used sampling techniques.

In this study, we focus on wind distributed (anemophilous) pollen. Anemophilous species tend to produce high amounts of pollen to increase the possibility of successful pollination. Wind distributed pollen grains are usually small, lightweight and have shapes favorable to wind capture and transport (Muller, 1979; Niklas, 1985). However, some gymnosperms, for example certain species of the Pinaceae family, have large to very large pollen, yet they are pollinated by wind. The distinct characteristics of different pollen types is essential for the detection and monitoring of pollen, which will be explained in more detail in Sect. 2.1. A more detailed description of the most abundant pollen types at our measurement site will be given in Sect. 2.3.

Pollen dispersed in the air does not only cause allergic symptoms in affected individuals when reaching the respiratory tract, it also contributes to the aerosol load

and affects weather and climate. Different studies have shown that atmospheric pollen can affect cloud properties by acting as cloud condensation (CCN) or ice nuclei (IN). Pollen was found to act as efficient ice nuclei in the condensation freezing mode (Diehl et al., 2001) and the immersion and contact freezing mode (Diehl et al., 2002; von Blohn et al., 2005). Diehl et al. (2002) showed that pollen can act as immersion and contact freezing nuclei at warmer temperatures than non-biological particles. Thus, the importance of pollen in cloud and precipitation processes is undeniable. Furthermore, ruptured pollen grains can affect climate and human health as well. The rupture of pollen grains has been observed in several laboratory experiments. Flowers from grasses and wind pollinating trees were exposed to a cycle of wetting, drying and wind disturbances and fragments of pollen grains down to sub-micrometer size were observed (Hughes et al., 2020; Miguel et al., 2006; Taylor et al., 2002, 2004). Taylor et al. (2004) found that birch pollen can rupture while still on the catkins and measured fragments of birch pollen grains ranging from 30 nm to 4 μ m in size. Those pollen fragments can persist much longer in the atmosphere than intact pollen due to their small size and lower falling velocity. In the atmosphere, those pollen fragments can act as CCN (Steiner et al., 2015; Wozniak et al., 2018). Moreover, small pollen fragments can penetrate deeper into the respiratory system and trigger more severe allergic reactions and asthma (Taylor and Jonsson, 2004).

2.1 Pollen monitoring

Pollen type and concentration are usually measured near ground, i.e. on roof-level. Worldwide at least 879 active stations continuously monitored the pollen prevalence in 2016 (Buters et al., 2018). The majority (over 600) of the stations are operating devices based on the Hirst principle, which will be introduced hereafter. The operation of this type of pollen trap and the analysis of the pollen samples are done manually. Thus, it requires human resources, is time consuming and near-real time measurements are not possible. Efforts have been made to develop automatic pollen detection techniques to reduce workload and enable pollen monitoring in near-real time. Those techniques use for instance image recognition such as the fully automated, image recognition-based pollen monitoring system, BAA500 (Oteros et al., 2015). Others make use of the fluorescence spectra of the particle like the Wideband Integrated Bioaerosol Sensor (WIBS) (Gabey et al., 2010; Savage et al., 2017) or the Real-Time Airborne Particle Identifier (Rapid-E) developed by Plair SA (Šaulienė et al., 2019).

The traditional and most commonly operated instruments to measure pollen type and concentration are Hirst-type volumetric spore samplers. The sampling prin-

ciple was developed by Hirst (1952). With this type of spore sampler it is possible to continuously collect pollen grains for 7 days with a time resolution of 2 hours. The sampling device consists of a rotatable sampler with a 14 mm x 2 mm orifice, a rain shield above the orifice and a wind vane. A pump draws air with a flow rate of 10 L min⁻¹ through the orifice into the sampling device. The large wind vane on the rotatable sampler ensures that the orifice is always oriented towards the wind. The sucked particles impact on an adhesive-coated plastic tape which is fixed on a rotating drum beneath the orifice. The tape is changed regularly and the pollen grains impacted on the tape are manually analyzed under the microscope. The number of counted spores on the sample tape surface is converted to pollen concentrations in pollen grains m⁻³ by using a conversion factor which considers the inlet air flow, sampling efficiency, surface area of the sampling tape and the number and area of analysed microscope fields. The pollen grains are classified according to their visual appearance. Depending on how different pollen grains within one family are, they can either be identified to genus level e.g. *Betula* or *Pinus* or only to the family level e.g. Poaceae. The Hirst-type pollen collector operated at our measurement site is shown in the right picture in Fig. 1.

Figure 1 Left: Vehmasmäki measurement station surrounded by broad-leaved and coniferous forest. Measurement instruments are located inside and on top of the measurement container and on the co-located mast. Right: Hirst-type pollen collector installed on top of the measurement container.

2.2 Observation overview

2.2.1 Measurement station

Finland offers ideal conditions for the observation of pollen as it is one of the most forested countries in Europe (FAO, 2020) with around 75% of Finland's total area being forestland (Lier et al., 2019). Air pollution levels are low (WHO, 2016), hence the amount of background aerosol is small. Another advantage of Finland as a location for pollen studies is the distance to large sources of depolarizing aerosols e.g. deserts, and therefore the lack of competing depolarizing aerosols. Figure 2 shows the location of the measurement site and the Corine land cover by the Copernicus Land Monitoring Service from the year 2018. Broad-leaved, coniferous and mixed forest are presented in green colours. The dominance of forest

Figure 2 Illustration of the Corine Land Cover 2018 © European Union, Copernicus Land Monitoring Service 2018, European Environment Agency (EEA) for Pan-Europe and the area surrounding the Vehmasmäki measurement site (inset). The complete legend can be accessed at Copernicus Land Monitoring Service (2018).

land in the Nordic countries is clearly apparent, while in Central and Southern Europe urban areas and cultivated land are strongly influencing the landscape.

The measurement station for pollen observations in the years 2016, 2019 and 2020 was located at Vehmasmäki (62°44' N, 27°33' E), a rural forest site around 18 km from Kuopio, Eastern Finland. This field station of the Finnish Meteorological Institute (FMI) has been established in 2012 and the operated lidar instrument belongs to the EARLINET network. The land cover around the measurement station (marked by a black triangle) is shown in the inset of Fig. 2. The station is mainly surrounded by broad-leaved, coniferous and mixed forest. Because of this location, far from major aerosol sources like deserts or big cities, Vehmasmäki is well suited for pollen observations. The measurement station and its surrounding can be seen in Fig. 1. Instruments are located inside and on top of a measurement container. Continuous remote-sensing observations are performed with a multiwavelength Raman polarization lidar Polly^{XT}, a Halo StreamLine Doppler lidar and a Vaisala CL51 ceilometer. Furthermore, a variety of in-situ instruments measure the aerosol situation on ground level. An optical particle sizer (OPS), Aerodynamic particle sizer (APS), Scanning mobility particle sizer (SMPS), Synchronized Hybrid Ambient Real-time Particulate Monitor (SHARP), Nephelometer and Aethalometer are operated permanently at the measurement site. Meteorological parameters such as temperature, relative humidity and wind speed and direction are measured in several heights between 2 and 300 m on a co-located mast (visible in Fig.1) and can deliver additional information about the state of the boundary layer.

2.2.2 Pollen observations in Vehmasmäki

23 different pollen types have been detected in Vehmasmäki during the years 2016, 2019 and 2020. Figure 3a shows the annual pollen concentration of the pollen families and genera detected in Vehmasmäki for the years 2016, 2019 and 2020 in blue, black and grey, respectively. Birch (*Betula*), pine (*Pinus*) and spruce (*Picea*) are the most abundant pollen genera in Vehmasmäki, followed by *Urtica* and the Poaceae family. The total number of detected pollen varies from year to year, so that the total pine pollen concentration, for instance, was slightly higher than the birch pollen concentration in 2016 and approximately similar in 2020. In 2019, however, significantly more birch pollen was detected. In Figure 3a the detected pollen types are ranked according to their mean total pollen concentration.

The main pollen seasons of the in Vehmasmäki detected pollen types are shown in Fig. 3b. The main pollen seasons, the time period during which most pollen of specific type is recorded, can be defined in various ways. Usually either the percentage of the annual pollen concentration or a certain threshold of the daily

Figure 3 Pollen genera and families detected in Vehmasmäki in the years 2016, 2019 and 2020. (a) Annual total pollen concentration of each pollen type for each year. (b) Pollen seasons of each pollen type determined using the 95% method (Goldberg et al., 1988). One-time pollen observations are marked by a dot.

pollen concentration is used to define start and end date of the pollen season (Jato et al., 2006). Here the main pollen seasons are defined using the 95% method described by Goldberg et al. (1988) and Andersen (1991). The beginning of the pollen season is defined as the first day when the sum of daily mean pollen concentrations reaches 2.5% of the annual concentration of the respective type. The end is the day when 97.5% of the annual total is reached. The pollen season therefore includes 95% of the annual total pollen concentration. Alternatively, the main pollen season is considered over when a period of dense occurrence was followed by 10 consecutive days with no detected pollen. The start of pollen seasons is strongly dependent on weather conditions like temperature and humidity (Stach et al., 2008; Zhang et al., 2015). Thus, the seasons of the individual pollen types vary each year and are here shown separately for the years 2016, 2019 and 2020 in blue, black and grey, respectively. The first pollens to be detected are usually from deciduous trees like alder (*Alnus*), poplar (*Populus*) and willow (*Salix*). Birch

(*Betula*) pollen can be observed throughout May. The season for coniferous pollen types, *Picea* and *Pinus*, is from the middle of May to June. Grass (Poaceae) and *Urtica* pollen seasons start in the middle of summer and last until autumn. Several pollen types, e.g. *Fraxinus*, Ericaceae, Asteraceae, *Plantago* and Cannabaceae have only been observed sporadically on individual days and are presented with dots.

2.3 Description of the most abundant pollen types

The detection of pollen types is based on the visual appearance of the grains. In the microscopical analysis, pollen grains are classified according to e.g. their size, apertures, which are areas in the pollen wall that significantly differ from their surrounding (Halbritter et al., 2018), the number of pores and the surface ornamentation i.e. surface features. These physical features of the pollen grains also matter when detecting pollen using optical remote sensing techniques, i.e. lidar. To illustrate the visual difference of the most common pollen types detected in Vehmasmäki, examples of local occurring pollen species of the most abundant pollen genera/families are presented in Table 1 and a description of their genus/family, their allergenicity and the visual appearance of the pollen grains is given.

Birch (*Betula*) pollen is the most allergenic tree pollen in northern, central and eastern Europe (D'Amato et al., 2007) and can cause severe allergic rhinitis and possibly even asthmatic symptoms (Biedermann et al., 2019; Schäppi et al., 1997). Birch trees are deciduous and belong to the Betulaceae family which, among others, also include Alder and Hazel. Species of the *Betula* genus native to northern Europe are for example Silver birch (*Betula pendula*), Downy birch (*Betula pubescens*) or Dwarf birch (*Betula nana*). Their pollen grains belong to the class of triporate pollen, which means they have three pores on their surface. In hydrated/fresh condition, the pollen grains are spherical and 20-30 μm in diameter. The round to oval pores have a diameter of 2-3 μm (Nilsson et al., 1977). The surface of birch pollen grains is rather smooth (Blackmore et al., 2003). When dry, the pollen grains become irregular shaped as the pollen wall folds onto itself to prevent further dehydration. This characteristic folding of pollen grains is known as harmomegathy (Katifori et al., 2010; Wodehouse, 1935). The pollen grains of the *Betula* genus show irregular infoldings with the interapertural area sunken in dry condition (Halbritter et al., 2018).

Pine (*Pinus*) and spruce (*Picea*) belong to the Pine family (Pinaceae). This family includes 12 genera (Simpson, 2010) of coniferous trees and bushes such as firs (*Abies*), larches (*Larix*), Douglas firs (*Pseudotsuga*) and cedars (*Cedrus*) besides pine and spruce. In Finland, pine and spruce make up 80% of the forest land, with

Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*) being the main tree species (Luke, 2020). Although being very abundant in Finland, Pinaceae pollen is only low allergenic. The pollen grains of Pinaceae species are quite big and possess air bladders, or sacci, which assist the pollen grains to be lifted and transported by wind. The sacci increase the surface area of the pollen grain without adding a lot of mass. Thereby they reduce the settling velocity and increase the dispersal distance (Schwendemann et al., 2007). The body (*corpus*) is circular to ellipsoidal and has a wrinkly, wart-like structure, while the bladders are rather smooth (Bykowska and Klimko, 2015; Grímsson and Zetter, 2011). The two most abundant coniferous pollen types in Finland, *Picea abies* and *Pinus sylvestris*, differ by their size. *Picea abies* pollen grains are very large with a diameter of 90-110 μm , while the grains of *Pinus sylvestris* pollen are 65-80 μm on their longest axis including the sacci (Nilsson et al., 1977). Furthermore, the sacci of *Pinus sylvestris* pollen are more protruding. When dry, the thin-walled area between the two sacci sinks in (Halbritter et al., 2018).

Urtica is a genus in the family Urticaceae, the family of flowering plants known as the nettle family. This family includes about 54 genera and 2,625 species of herbs, shrubs and small trees. Representative species in Finland are the common nettle or stinging nettle (*Urtica dioica*) and the burning nettle (*Urtica urens*), both are herbaceous plants of the *Urtica* genus. The pollen of this genus is considered poorly to moderate allergenic (Tiotiu et al., 2016). Their pollen grains are small (15-20 μm), oblate-spheroidal to spheroidal and have 3-4 circular pores (Nilsson et al., 1977). Dry pollen grains are irregularly infolded.

Poaceae is the family of flowering plants known as grasses. This plant family is one of the largest with approximately 780 genera and 12,000 species (Christenhusz and Byng, 2016). Species of this family are among the most abundant and widely spread plants on Earth. Poaceae pollen is among the leading aeroallergens worldwide and considered the main cause of pollen allergy in most developed countries (D'Amato et al., 2007; García-Mozo, 2017). It can cause severe symptoms of allergic rhinitis, asthma, and conjunctivitis (García-Mozo, 2017). The pollen grains of Poaceae species are typically spheroidal in shape. When dry the pollen grains have irregular infoldings. Poaceae pollen belong to the class of ulcerate pollen, which means the pollen grains have a single distally located ulcer, a more or less circular pore-like aperture. On their surface, Poaceae pollen grains have nano-sized sculpture elements (Halbritter et al., 2018). The size range of pollen grains within the Poaceae family is highly variable (Joly et al., 2007; Salgado-Labouriau and Rinaldi, 1990) and can range from small e.g. *Poa annua* (annual bluegrass) to large pollen grains e.g. *Triticum aestivum* (Common wheat). Thereby large pollen grains usually belong to cultivated species (Erdtman, 1986).

Taxa (Genus/Family)	Pollen class	Diameter (longest axis)	Shape	Micrograph	Exine surface micrograph	Reference
<i>Betula pendula</i> (<i>Betula</i> /Betulaceae)	porate	21-25 μm	spheroidal			Halbritter et al. (2020a)
<i>Pinus sylvestris</i> (<i>Pinus</i> /Pinaceae)	saccate	65-80 μm^* (sacci included)	saccate			Halbritter and Heigl (2020b)
<i>Picea abies</i> (<i>Picea</i> /Pinaceae)	saccate	90-110 μm^* (sacci included)	saccate			Halbritter and Heigl (2020a)
<i>Urtica dioica</i> (<i>Urtica</i> /Urticaceae)	porate	15-20 μm^*	spheroidal			Halbritter et al. (2020b)
<i>Poa annua</i> (<i>Poa</i> /Poaceae)	ulcerate	10-25 μm	spheroidal			Halbritter and Schneider (2015)

Table 1 Pollen morphology of representatives of the five most abundant pollen genera/families detected in Vehmasmäki. Exine surface micrographs of saccate pollen are from the corpus of the pollen grain. Characteristics are taken from the Paldat website (Paldat, 2000), individual references are given in the table.

* taken from Nilsson et al. (1977).

3 Lidar methodology

LIDAR, the acronym for "Light Detection And Ranging", is an active remote sensing instrument, which allows the measurement of vertical profiles of atmospheric parameters such as temperature, humidity and wind, aerosol and cloud optical properties and the vertical distribution of gases. Laser pulses at a number of selected wavelengths and polarization states are emitted into the atmosphere, where the laser light is scattered and absorbed by aerosol particles or the molecules of atmospheric gases. The part of the emitted light that is scattered back to the instrument is received by a telescope. Apart from the distance of the backscattering object, other properties can be derived. The properties of the atmospheric constituents are measured in ambient conditions, which means the observations are not affected by the measurements itself. Another advantage of lidar remote sensing is the good vertical and time resolution of the measurements compared to other profiling methods for instance soundings or measurements aboard aircraft.

Since the first attempts to use light for determining air density profiles in the upper atmosphere in the 1930s (Johnson et al., 1939; Synge, 1930) and the first applications using laser as light source for atmospheric lidar observation by Fiocco and Smullin (1963), major progress in the field of lidar technology has been achieved. Lidar systems have been continuously evolved and various types of lidars, specialized for different applications have been developed and are nowadays deployed on ground (ground-based) as well as on board of aircraft (airborne) and satellites (spaceborne). Two types of lidar systems, in-elastic (Raman) lidars and Doppler wind lidars are introduced in Section 3.5 and Section 3.6. Representatives of the mentioned lidar types, the multiwavelength Raman lidar Polly^{XT} and the Halo StreamLine Doppler lidar were used in the presented study. First, the lidar principle and lidar equation are introduced (Sect. 3.1) and derived optical parameters are explained.

3.1 Lidar principle and equations

Lidar systems emit monochromatic and coherent laser pulses of a certain polarization state into the atmosphere. In the atmosphere, the laser pulses interact with molecules and particles through scattering and absorption. The pulses that are backscattered at an angle of 180° to the lidar are collected by a telescope. From the time interval t between emission of laser pulse and detection of the backscattered photon, the distance, $R = tc/2$, can be determined. The laser pulse travels at the speed of light c to the backscattering object and back, which explains the division by 2. The following equation describes the received signal power P from the range R at the emission wavelength λ (Wandinger, 2005a):

$$P_{\lambda}(R) = P_0 C_{\lambda}^s \frac{O(R)}{R^2} \beta_{\lambda}(R) \exp \left[-2 \int_0^R \alpha_{\lambda}(r) dr \right] + P_{bg}. \quad (3.1)$$

P_0 is the power of the emitted laser pulse at the wavelength λ . The constant C_{λ}^s summarizes height independent system parameters such as receiver optics and detectors efficiency, telescope area and pulse duration. $O(R)$ describes the overlap between laser beam and receiver field-of-view. This parameter varies between 0 close to the instrument, which means no overlap, and 1, meaning full overlap. The volume backscatter coefficient $\beta_{\lambda}(R)$ describes the amount of light backscattered at an angle of 180° . It is defined as the product of the number concentration of scatterers of kind j and its differential scattering cross section $d\sigma_{j,sca}/d\Omega$ in backward direction ($\Theta = \pi$) at the wavelength λ summed over all kinds of scatterers. Since light is backscattered at molecules and particles, the backscatter coefficient can be written as sum of particle and molecular backscatter coefficient:

$$\beta_{\lambda}(R) = \sum_j N_j(R) \frac{d\sigma_{j,sca}}{d\Omega}(\pi, \lambda) = \beta_{\lambda}^{par}(R) + \beta_{\lambda}^{mol}(R). \quad (3.2)$$

The exponential term in Eq. 3.1 describes the two-way transmission of the light from the instrument to the distance R and back, therefore a factor of two is applied. The attenuation of light, i.e. scattering and absorption on molecules and particles, depends on the length of the light path and the volume extinction coefficient $\alpha_{\lambda}(R)$ which is defined as:

$$\alpha_{\lambda}(R) = \alpha_{\lambda}^{par}(R) + \alpha_{\lambda}^{mol}(R). \quad (3.3)$$

The detected signal always comprises a background signal P_{bg} , which can for example originate from direct or scattered sunlight or artificial light sources. The receiver telescope area can be seen as part of the surface of the sphere with radius R that surrounds the scattering volume, the received signal $P_{\lambda}(R)$ is therefore inversely proportional to R^2 . The signal intensity decreases with increasing distance from the lidar. Thus, the received signal multiplied by R^2 , known as the range-corrected signal, is commonly used for aerosol studies.

Equation 3.1 describes the received signal for elastic scattering, which means there is no energy transfer between photon and particle or molecule during the scattering process and the received wavelength is identical to the emitted wavelength. Raman lidar systems can additionally record the inelastic (Raman) backscattered signal from Raman-active molecules e.g. nitrogen, oxygen or water vapour. During the inelastic scattering process, energy is exchanged between the photon and the interacting molecule. This leads to a change of the vibrational-rotational energy

state of the molecule and a shift of the wavelength of the scattered photon. The wavelength shift is characteristic for the backscattering molecule. For instance, the inelastic scattering by nitrogen molecules causes a wavelength shift for the initial wavelengths 355 nm and 532 nm to the Raman wavelengths 387 nm and 607 nm, respectively. The characteristic Raman-shifted wavelength for inelastic scattering on water vapour molecules is 407 nm for the initial wavelength of 355 nm. The lidar equation for inelastic scattering at the Raman-shifted wavelength λ_{Ra} can be described as (Wandinger, 2005b):

$$P_{\lambda_{Ra}}(R) = P_0 C_{\lambda_{Ra}}^s \frac{O(R)}{R^2} \beta_{\lambda_{Ra}}(R) \exp \left[- \int_0^R (\alpha_{\lambda_0}(r) + \alpha_{\lambda_{Ra}}(r)) dr \right], \quad (3.4)$$

with the Raman backscatter coefficient

$$\beta_{\lambda_{Ra}}(R) = N_{Ra}(R) \frac{d\sigma_{Ra}}{d\Omega}(\pi, \lambda_{Ra}). \quad (3.5)$$

Unlike in the equation for elastic scattering (Eq. 3.1), the transmission term in the Raman lidar equation (Eq. 3.4) considers the extinction on the way to the scattering volume at the initial wavelength α_{λ_0} and from the scattering volume back to the lidar at the Raman wavelength $\alpha_{\lambda_{Ra}}$. Both extinction coefficients consider again the extinction at particles and molecules (according to Eq. 3.3).

The Raman backscatter coefficient $\beta_{\lambda_{Ra}}(R)$ is defined by the molecule number density of the Raman-active gas N_{Ra} and the differential Raman cross section of the gas in backward direction $d\sigma(\pi)/d\Omega$. Using temperature and pressure profiles from radio soundings, models, e.g. GDAS (Global Data Assimilation System, GDAS (2021)) or the standard atmosphere, N_{Ra} can be calculated.

The particle extinction coefficient can directly be derived from the inelastic (Raman) backscattered signal (Eq. 3.4), as (Ansmann et al., 1990):

$$\alpha_{\lambda_0}^{par}(R) = \frac{\frac{d}{dR} \ln \left[\frac{N_{Ra}(R)O(R)}{R^2 P_{\lambda_{Ra}}(R)} \right] - \alpha_{\lambda_0}^{mol}(R) - \alpha_{\lambda_{Ra}}^{mol}(R)}{1 + \left(\frac{\lambda_0}{\lambda_{Ra}} \right)^{\mathring{A}_{ext}}}, \quad (3.6)$$

with $\mathring{A}_{ext}$ being the extinction-related Ångström exponent (see Sect. 3.4), which describes the wavelength dependence of the extinction coefficient of particles. The particle backscatter coefficient can be calculated using both the inelastic and elastic

backscattered signal according to (Ansmann et al., 1992) as:

$$\beta_{\lambda_0}^{par}(R) = -\beta_{\lambda_0}^{mol}(R) + \left[\beta_{\lambda_0}^{par}(R_0) + \beta_{\lambda_0}^{mol}(R_0) \right] \cdot \frac{P_{\lambda_{Ra}}(R_0) P_{\lambda_0}(R) N_{Ra}(R)}{P_{\lambda_0}(R_0) P_{\lambda_{Ra}}(R) N_{Ra}(R_0)} \cdot \frac{\exp \left[-\int_{R_0}^R \left(\alpha_{\lambda_{Ra}}^{par}(r) + \alpha_{\lambda_{Ra}}^{mol}(r) \right) dr \right]}{\exp \left[-\int_{R_0}^R \left(\alpha_{\lambda_0}^{par}(r) + \alpha_{\lambda_0}^{mol}(r) \right) dr \right]}, \quad (3.7)$$

with R_0 being a selected reference height. The reference height is usually selected as a presumably aerosol-free region in order to calibrate the lidar signal with a known atmospheric target. The particle backscatter coefficient $\beta_{\lambda_0}^{par}(R_0)$ in this height is often assumed to be zero or a very small fraction of the molecular backscatter coefficient.

The derived particle backscatter- and extinction coefficients are *extensive* optical properties. This means they depend on the nature of the particle (shape, composition) as well as on the amount of backscattering particles in the atmosphere. They give an indication of the amount of backscattered or extinguished light. Furthermore, particle backscatter- and extinction coefficients are used to calculate *intensive* optical properties, e.g. depolarization ratios (Sect. 3.2), lidar ratios (Sect. 3.3) and Ångström exponents (Sect. 3.4). *Intensive* optical properties do not depend on the amount but only the nature of the particle and can therefore be used to characterize particles and distinguish different aerosol types.

For most Raman lidars, the described retrieval of particle backscatter- and extinction coefficients is limited to nighttime measurement as the background noise in the Raman channels caused by daylight is usually too high to apply the Raman method. In this case, the Klett-Fernald method (Fernald, 1984; Klett, 1981) has to be applied to solve the lidar equation. When not exploiting the Raman signal, the lidar equation (3.1) contains two unknown parameters, the backscatter- and extinction coefficient. As no additional signal from inelastic backscattered light is detected, backscatter- and extinction coefficient cannot be determined independently anymore. The so-called lidar ratio LR , the ratio of extinction- to backscatter coefficient (see Sect. 3.3), has to be assumed to solve the equation. Using an estimated particle backscatter coefficient $\beta_{\lambda}^{par}(R_0)$ at the reference height R_0 , the molecular backscatter coefficient $\beta_{\lambda}^{mol}(R)$ and the molecular lidar ratio LR^{mol} , the particle backscatter coefficient can then be derived as:

$$\beta_{\lambda}^{par}(R) = -\beta_{\lambda}^{mol}(R) + \frac{A_{\lambda}(R_0, R)}{B_{\lambda}(R_0) - 2LR_{\lambda}^{par} \int_{R_0}^R A_{\lambda}(R_0, r) dr}, \quad (3.8)$$

with

$$A_\lambda(R_0, R) = P_\lambda(R) R^2 \exp \left[-2 \left(LR_\lambda^{par} - LR^{mol} \right) \int_{R_0}^R \beta_\lambda^{mol}(r) dr \right] \quad (3.9)$$

and

$$B_\lambda(R_0) = \frac{P_\lambda(R_0) R_0^2}{\beta_\lambda^{par}(R_0) + \beta_\lambda^{mol}(R_0)}. \quad (3.10)$$

The particle extinction coefficient can be obtained through the relation of lidar ratio and backscatter coefficient. Because of the assumption of a lidar ratio, profiles retrieved with the Klett-Fernald method are prone to more systematic errors (Sasano et al., 1985) than profiles retrieved with the Raman method, while those are stronger affected by noise.

3.2 Depolarization measurements

The light emitted by the lidar instrument is linearly polarized which means the orientation of the electric field vector is fixed. When interacting with atmospheric components, the polarization state of the light can change. The backscattered light then contains a cross-polarized component $P_\lambda^\perp$ in addition to the light that is polarized in the same plane as the emitted light, the parallel polarized component $P_\lambda^\parallel$. Lidars with polarization measurement capacities can detect these components of the backscattered light and the lidar equation (Eq. 3.1) can be expressed as:

$$P_\lambda^\parallel(R) = P_0 C_\lambda^{s\parallel} \frac{O_\lambda(R)^\parallel}{R^2} \beta_\lambda^\parallel(R) \exp \left[-2 \int_0^R \alpha_\lambda^\parallel(r) dr \right] \quad (3.11)$$

and

$$P_\lambda^\perp(R) = P_0 C_\lambda^{s\perp} \frac{O_\lambda(R)^\perp}{R^2} \beta_\lambda^\perp(R) \exp \left[- \int_0^R \left[\alpha_\lambda^\parallel(r) + \alpha_\lambda^\perp(r) \right] dr \right], \quad (3.12)$$

with the superscript $\parallel$ and $\perp$ denoting the parallel and perpendicular plane of polarization with respect to the polarization plane of the emitted laser light, respectively. It is usually assumed, that the atmospheric extinction does not depend on the polarization state and that parallel and perpendicular extinction are equal. The transmission terms of the parallel and perpendicular component of the detected signal are therefore usually assumed identical. Using preceding equations,

the volume depolarization ratio can be calculated as the ratio of perpendicular to parallel backscatter component:

$$\delta_{\lambda}^{vol}(R) = \frac{\beta_{\lambda}^{\perp}(R)}{\beta_{\lambda}^{\parallel}(R)} = \frac{\beta_{\lambda}^{mol,\perp}(R) + \beta_{\lambda}^{par,\perp}(R)}{\beta_{\lambda}^{mol,\parallel}(R) + \beta_{\lambda}^{par,\parallel}(R)}. \quad (3.13)$$

Both backscatter coefficients include the part of the light backscattered by molecules and particles. The ratio of perpendicular to parallel molecular or particle backscatter coefficients is called molecular depolarization ratio $\delta_{\lambda}^{mol}(R)$ or particle depolarization ratio $\delta_{\lambda}^{par}(R)$, respectively.

The particle depolarization ratio is an intensive quantity and allows the determination of the scatterers sphericity. Non-spherical particles cause a depolarization of the backscattered light because the orientation of the polarization changes during the scattering within the particle or on its edges. Spherical particles, however, do not cause a depolarization of the light and the depolarization ratio is zero. Aerosol particles can thus be characterized based on their shape (Groß et al., 2011; Haarig et al., 2018), which makes the depolarization ratio a valuable quantity for aerosol classification. Furthermore, the depolarization ratio can be used to separate the contribution of spherical and non-spherical particles to the lidar signal following a method described by Tesche et al. (2009a). Table 2 gives examples of particle depolarization ratios observed in previous studies using lidar measurements to illustrate the difference between different aerosol types.

Table 2 Examples of depolarization ratios for different aerosol types.

Aerosol type	$\delta_{355}^{par}(\%)$	$\delta_{532}^{par}(\%)$	$\delta_{1064}^{par}(\%)$	Reference
Saharan dust	26±6	31±3	-	Freudenthaler et al. (2009)
Smoke (tropos.)	2±4	3±2	1±0.8	Haarig et al. (2018)
Marine (wet)	≤3	≤2	≤2	Haarig et al. (2017)
Marine (dry)	11.5±8.2	14.8±3.5	9.9±1.1	Haarig et al. (2017)
Volcanic	35-38	35-38	-	Groß et al. (2012)

3.3 Lidar ratio

The ratio of extinction to backscatter coefficient at the considered wavelength λ is called lidar ratio:

$$LR_{\lambda}(R) = \frac{\alpha_{\lambda}(R)}{\beta_{\lambda}(R)}. \quad (3.14)$$

The LR is a necessary assumption for retrieving backscatter profiles using the Klett-Fernald method but can also be used for aerosol characterization when using the Raman method and obtaining backscatter- and extinction profiles independently. It has be differentiated between molecular lidar ratio LR^{mol} and particle lidar ratio LR^{par} . While the molecular LR is well-defined for air ($LR^{mol} = 8\pi/3$ sr), the particle LR depends on size distribution, shape, humidity and chemical composition of the backscattering particles. The particle lidar ratio is thus often used to characterize and discriminate between different aerosol types. Table 3 shows examples of lidar ratios at 355 and 532 nm for different aerosol types.

Table 3 Examples of lidar ratios for different aerosol types.

Aerosol type	LR_{355}^{par} (sr)	LR_{532}^{par} (sr)	Reference
Saharan dust	53±10	54±10	Tesche et al. (2011a)
Smoke	87±17	79±17	Tesche et al. (2011b)
Marine (wet)	19±5	23±2	Haarig et al. (2017)
Marine (dry)	27±6	25±3	Haarig et al. (2017)
Continental polluted	58±12	53±11	Müller et al. (2007)
Volcanic	50-60	45-55	Groß et al. (2012)

3.4 Ångström exponent

The measurement of optical signals at multiple wavelengths allows the determination of the Ångström exponent $\text{\AA}$. The Ångström exponent describes the ratio of a parameter x at two wavelengths λ_1 and λ_2 as a function of the ratio of these wavelengths (Ångström, 1929):

$$\frac{x_{\lambda_1}}{x_{\lambda_2}} = \frac{\lambda_2^{\text{\AA}_{\lambda_1/\lambda_2}^x}}{\lambda_1^{\text{\AA}_{\lambda_1/\lambda_2}^x}}, \quad \text{\AA}_{\lambda_1/\lambda_2}^x = \frac{\ln[x_{\lambda_1}/x_{\lambda_2}]}{\ln[\lambda_2/\lambda_1]}. \quad (3.15)$$

It describes the spectral dependence of the parameter x , which can be, for example, the backscatter or extinction coefficient (Ansmann and Müller, 2005). The scattering on large particles is almost wavelength independent, the extinction-related Ångström exponent is therefore approximately zero. Small particles, on the other hand, correspond to a large $\text{\AA}$ as the scattering is strongly dependent on the wavelength. Hence, the Ångström exponent is often used to estimate the dominating size range of the scattering particles. Examples for Ångström exponents determined in previous studies are given in Table 4. However, values can

vary dependent on the aerosol type, source and the mixture of aerosols in the atmosphere. For example, also negative Ångström exponents have been reported for Saharan dust in a several studies (Bohlmann et al., 2018; Rittmeister et al., 2017; Veselovskii et al., 2016).

Table 4 Examples of extinction (α) and backscatter (β) related Ångström exponents for different aerosol types.

Aerosol type	$\text{\AA}^\alpha_{355/532}$	$\text{\AA}^\beta_{355/532}$	$\text{\AA}^\beta_{532/1064}$	Reference
Saharan dust	0.06 ± 0.21	0.19 ± 0.20	0.28 ± 0.16	Tesche et al. (2009b)
Smoke	1.15 ± 0.28	0.90 ± 0.26	1.06 ± 0.65	Tesche et al. (2011b)
Marine aerosol	0.3 ± 0.1	-	-	Müller et al. (2007)
Continental polluted	1.4 ± 0.5	-	1.3 ± 0.5	Müller et al. (2007)
Volcanic	-	0.6-0.8	-	Sicard et al. (2012)

3.5 Multiwavelength Raman polarization lidar Polly^{XT}

Since 2012, a multiwavelength Raman polarization lidar (Polly^{XT}) is permanently operated at the measurement site in Vehmasmäki (Kuopio). Polly^{XT} (POrtabLe Lidar sYstem, eXTended version) has been developed at the Leibniz Institute for Tropospheric Research (Althausen et al., 2009; Engelmann et al., 2016) and is part of the PollyNET network (Baars et al., 2016).

Figure 4 shows a schematic illustration of the Polly^{XT} setup. Polly^{XT} lidar systems operate a Nd:YAG laser which emits light pulses at 1064 nm with a repetition frequency of 20 Hz. By doubling and tripling of this frequency in the second and third harmonic oscillator (SHG and THG), light pulses with a wavelength of 532 and 355 nm are generated. The light is directed through a Brewster-cut Glan laser polarizer to ensure linear polarization of the emitted light. For quality control, parts of the light are directed to a power meter which measures the UV component and allows the monitoring of the conversion efficiency. Two mirrors direct the light upwards. The light passes through a shutter which is used to interrupt the light path in case of airplane overflights. Finally, the light beam is enlarged to a diameter of 45 mm by a beam expander before being emitted into the atmosphere. The receiver unit consists of a Newtonian telescope with a 30 cm primary mirror and a secondary mirror which directs the light through a pinhole to the detection unit. A polarizer mounted in front of the pinhole allows the calibra-

tion of the depolarization measurements (see Sect. 3.5.2). Dichroic beam splitters separate the detected light by wavelength and mirrors direct the light to the respective photo multiplier tube (PMT), which are connected with the data acquisition unit. Photons are counted at the three emitted wavelengths (355, 532 and 1064 nm) and the Raman-shifted wavelengths (387, 407 and 607 nm) and the cross-polarized light at 532 and 355 nm. The signal is stored with a temporal resolution of 30 s. To avoid specular reflections from horizontally aligned ice crystals, the whole system is tilted by 5° (Seifert et al., 2010).

The Polly^{XT} system operated in Vehmasmäki was updated in 2017. Before, Polly^{XT}-FMI had just one depolarization channel at 532 nm. The new version

Figure 4 Schematic illustration of the optical setup of Polly^{XT}. The lower part displays the front view, while the upper part shows the top view including the additional near-range receiver. Adapted from Engelmann et al. (2016), a more detailed illustration and description can be found therein.

comprises a second depolarization channel at 355 nm and a near-field unit (see Sect. 3.5.1). Furthermore, the vertical resolution was improved from 30 to 7.5 m. Results used in **Papers I to III** utilized measurements with the old Polly^{XT} system without near-field unit and second depolarization channel. Measurements for **Paper IV** have been done with the new setup.

3.5.1 Overlap effect and near-field unit

Polly^{XT} is a bi-axial systems, which means emission and detection unit are not on the same axis and the overlap of laser beam and telescope field-of-view (FOV) needs to be considered. Using a camera synchronized to the laser trigger (see Fig. 4), it is possible to monitor and adjust the overlap of laser beam and the FOV of the telescope. However, laser beam and telescope FOV do not overlap close to the lidar, and thus the minimal height of usable profiles is limited. Full overlap of the primary (far-field) receiver and the laser beam is reached at around 800-900 m (Engelmann et al., 2016). To decrease the minimal measurement height, i.e. the height of full overlap of laser beam and receiver FOV, a secondary smaller telescope with a wider field-of-view (2.2 mrad) and close distance to the emission axis has been added to the upgraded version of Polly^{XT}-FMI (see top view, Fig. 4). Near-field signals are detected at 355 and 532 nm and the Raman-shifted wavelengths at 387 and 607 nm. Due to this setup it is possible to detect profiles down to approximately 120 m instead of the previous limitation to around 800 m (Engelmann et al., 2016).

3.5.2 Polly^{XT} depolarization measurements

Polly^{XT} comprises polarization capabilities. Thus, light with a polarization orientation differing from the emitted light can be recognized. The cross-polarized signal is detected at 532 nm and with the upgraded version of Polly^{XT} also at 355 nm. However, only the total backscattered and not the parallel polarized signal is detected at these wavelengths. Equation 3.13 can therefore not be used directly to calculate the volume depolarization ratio. Furthermore, the dichroic beam splitters show different transmissions for cross- and parallel polarized light which leads to different transmission efficiencies for cross and parallel polarized light in the detection channel (Engelmann et al., 2016). Consequently, depolarization measurements need to be calibrated to ensure correct depolarization ratios. For Polly^{XT} systems, the so-called $\Delta 90^\circ$ calibration method (Freudenthaler, 2016; Freudenthaler et al., 2009) is usually applied. Calibration measurements are performed three times a day by rotating a polarizer under $+45^\circ$ and -45° with respect to the laser

polarization plane into the light path and measuring the ratio of cross-polarized to total signal δ_λ^* for two subsequent measurements (Engelmann et al., 2016). The calibration constant V_λ^* is then defined according to Freudenthaler et al. (2009) as:

$$V_\lambda^* = \frac{1 + D_\lambda^{tot}}{1 + D_\lambda^c} \sqrt{\delta_{\lambda,+45^\circ}^*(R) \cdot \delta_{\lambda,-45^\circ}^*(R)}, \quad (3.16)$$

with D_λ^{tot} and D_λ^c being the transmission ratio of the total channel and the cross-polarized channel, respectively. The transmission ratios depend on the installed beam splitters and are different for each Polly system. Having the ratio of cross-polarized and total signal without polarizer in the light path $\delta_{0^\circ}^*$, the calibration constant V_λ^* and the transmission ratios D_λ^{tot} and D_λ^c , the volume linear depolarization ratio is given by (Engelmann et al., 2016):

$$\delta_\lambda^{vol} = \frac{1 - \delta_{\lambda,0^\circ}^*(R)/V_\lambda^*}{D_\lambda^{tot} \cdot \delta_{\lambda,0^\circ}^*(R)/V_\lambda^* - D_\lambda^c}. \quad (3.17)$$

The particle depolarization ratio can then be calculated using the volume depolarization ratio, the molecular depolarization ratio δ_λ^{mol} and the particle and molecular backscatter coefficients, β_λ^{par} and β_λ^{mol} , as:

$$\delta_\lambda^{par}(R) = [\delta_\lambda^{vol}(R) + 1] \left(\frac{\beta_\lambda^{mol}(R) [\delta_\lambda^{mol}(R) - \delta_\lambda^{vol}(R)]}{\beta_\lambda^{par}(R) [1 + \delta_\lambda^{mol}(R)]} + 1 \right)^{-1} - 1. \quad (3.18)$$

3.6 Halo Doppler lidar

Another lidar system permanently operated at our measurement site is a Halo Photonics StreamLine Pro Doppler lidar (Pearson et al., 2009). Doppler lidars make use of the frequency shift of moving particles (Doppler effect) in the atmosphere to measure wind and turbulence with high spatial and temporal resolution. Doppler lidars are therefore frequently used to study wind profiles (e.g. Tuononen et al. (2017)) and boundary layer evolution (e.g. Manninen et al. (2018)) and are applied in the wind energy sector.

Halo StreamLine lidars emit linearly polarized light pulses at 1565 nm with a pulse repetition frequency of 10 or 15 kHz, depending on the lidar version. These lidars can be operated in scanning or vertically pointing mode which enables three-dimensional mapping of turbulent flows as well as the height- and time-resolved measurement of the vertical velocity. Halo Doppler lidars are equipped with a heterodyne detector. The optical path is constructed with fiber-optic components.

Emission and detection of the light pulses take place on the same axis, overlap effects are therefore omitted and profiles can be determined from a minimum range of 90 m with a spatial resolution of 30 m. In addition to the normal receiver channel, a cross-polar receiver channel with a fiber-optic polarizer can be incorporated through a fiber-optic switch. Co- and cross-polarized signals can thus be detected consecutively in vertically pointing mode. The integration time can be changed according to location and application and may vary from 1 to 30 s.

Halo StreamLine lidars provide the radial Doppler velocity and the signal-to-noise ratio (SNR) along the beam direction. After post-processing the SNR according to Vakkari et al. (2019), the attenuated backscatter β can be calculated from the SNR as:

$$\beta(R) = A \frac{SNR(R)}{T_f(R)}, \quad (3.19)$$

with R being the range from the instrument, A comprising system-specific constants (e.g. beam energy and receiver bandwidth) and the telescope focus function $T_f(R)$ (Pentikäinen et al., 2020).

The measurement of co- and cross polarized backscattered signals allows the determination of the depolarization ratio. Unlike Polly^{XT}, Halo StreamLine Doppler lidars lack a calibration unit for depolarization measurements nor allow the addition of a calibrator. Thus, the depolarization ratio has to be calibrated using the incomplete extinction, or bleed-through, at liquid cloud base. At liquid cloud base, the depolarization ratio should be zero as single scattering on spherical cloud droplets does not polarize the backscattered light. Non-zero depolarization ratios at liquid cloud base is therefore an indicator of bleed-through and can be used for calibration. Considering the observed bleed-through B , the depolarization ratio δ can be determined as:

$$\delta = \frac{SNR_{cross} - B \cdot SNR_{co}}{SNR_{co}}, \quad (3.20)$$

using the co- and cross-polarized signal-to-noise ratio, SNR_{co} and SNR_{cross} , respectively. The bleed-through correction and determination of the depolarization ratios using Halo StreamLine Doppler lidars is the topic of **Paper III** and will be discussed in detail therein.

4 Main results

In this section the main results of the original publications are presented. For more detailed results please refer to corresponding publication (**Paper I-IV**).

4.1 Ability to detect pollen with lidar

In **Paper I**, we explored the capabilities of ground-based lidar systems to detect pollen in the atmosphere. Up to now, there is no comprehensive and validated information about optical properties of atmospheric pollen in the atmosphere. To include pollen as aerosol type in classification schemes and provide vertical information about pollen concentration, more data is needed. During our first pollen campaign in 2016 at Vehmasmäki, Finland, we combined measurements of the multiwavelength Raman polarization lidar Polly^{XT} and a Hirst-type pollen collector to link lidar observations in the vertical column and observed pollen concentrations at ground level. Figure 5 shows the observations from 5 to 15 May. During this period high concentrations of birch pollen were detected (Fig. 5a), while a relatively large aerosol load was observed within the boundary layer (Fig. 5b). Two

Figure 5 Overview of the intense pollen period in May 2016. (a) Pollen concentration measured by Hirst-type pollen sampler, (b) range-corrected signal at 1064 nm and (c) volume depolarization ratio at 532 nm obtained by Polly^{XT} Raman polarization lidar. Figure adapted from Paper I © 2019 Atmospheric Chemistry and Physics.

periods can be identified considering the volume depolarization ratio (Fig. 5c) and observed pollen types. From 5 to 9 May birch pollen was dominating at ground level. The observed volume depolarization ratio of airborne particles was small, while during the second period from 12 to 15 May, depolarization ratios were clearly higher and a considerable amount of spruce pollen was detected in addition to birch. The higher depolarization ratios during the second period were likely caused by the highly non-spherical shape of the spruce pollen grains during the second period. The measurements from both periods have been analysed to retrieve the optical properties of the pollen within determined layers. Figure 6 shows the mean values of lidar ratio at 532 nm and backscatter-related Ångström exponent at 532–1064 nm against the particle depolarization ratio at 532 nm. The mean lidar ratio at 532 nm ranges from 31 sr to 74 sr, with mean values of 52 ± 12 sr for the first period (birch) and 62 ± 10 sr for the second period (birch-spruce mixture). These values are characteristic for other aerosol types such as dust and dust-smoke

Figure 6 Mean values of (a) lidar- vs. particle depolarization ratio at 532 nm and (b) backscatter-related Ångström exponent at 532–1064 nm vs. particle depolarization ratio at 532 nm in the pollen layers observed from 5 to 10 May (period 1, green) and from 12 to 15 May (period 2, black). Error bars denote the standard deviation. Marker size denotes the pollen concentration. Solid dots indicate Raman retrievals and dotted markers the Klett solution. Figure from Paper I © 2019 Atmospheric Chemistry and Physics.

mixtures (Groß et al., 2011; Tesche et al., 2011a), hence the lidar ratio alone is not a suitable parameter for the identification of pollen. The depolarization ratio, on the other hand, is clearly different in the two periods.

During the birch dominated period (period 1) the mean PDR at 532 nm is $10 \pm 6\%$, while it is $26 \pm 7\%$ during the second period when spruce was observed in addition to birch. These depolarization ratios are in the same range as values for dust and biomass burning aerosol mixtures (Tesche et al., 2011a) or dust and marine aerosol mixtures (Groß et al., 2011). Thus, backward trajectories of observed air masses have been analyzed to ensure the absence of other depolarizing aerosol. The backscatter-related Ångström exponent has a tendency towards smaller values with increasing depolarization ratio during both periods and the average Ångström exponent is smaller in the second period when large spruce pollen was observed. However, in both periods the detected Ångström exponent is rather large considering the large size of pollen grains. The backscattering is most likely dominated by fine-mode (non-pollen) background aerosols. Thus, we do not measure the optical properties of pure pollen but rather the mixture of background aerosol and pollen. Therefore, the Ångström exponent can only be used for pollen detection under the same or similar background conditions.

This first pollen campaign demonstrated the potential of lidar measurements, especially depolarization ratio measurements, to detect the presence of pollen in the atmosphere. Observed depolarization ratios are similar to typical values for dusty mixtures. As atmospheric pollen is not included in current aerosol classification schemes, it is likely misclassified as dusty mixtures. More measurements and auxiliary data is needed to improve the classification of pollen and certain challenges need to be addressed. First, the minimum height of lidar observations needs to be as low as possible. It can be assumed that most pollen is located close to the pollen source, near the ground. The full overlap height of the operated lidar system is at around 700 m which is rather high. A lower full overlap height and therefore lower profiles would provide more conclusive results. Second, the contribution of other aerosols such as anthropogenic pollution needs to be determined to separate the effect of pollen and background aerosol on the observed profiles using established separation methodologies for example by Tesche et al. (2009b). Moreover, the contribution of different pollen types needs to be determined to get characteristic values for specific pollen types. For this purpose, measurement with only one pollen type are needed.

4.2 Depolarization ratio of pure pollen

Using lidar remote sensing we always measure the optical properties of the mixture of different aerosols in the atmosphere. Pollen is mixed with background aerosols and the optical properties of specific pollen types cannot be measured directly. Tesche et al. (2009a) provides a method to separate backscatter profiles of depolarizing and non-depolarizing aerosols based on the difference of the depolarization ratio. This method could also be used to estimate the contribution of pollen to the lidar signal. However, the method requires the knowledge of the depolarization ratio of pure pollen, which is the desired property. We therefore developed an algorithm to estimate the depolarization ratio of pure pollen based on lidar measurements which is presented in **Paper II**.

For this method, backscatter measurements at two or more wavelengths are necessary as the Ångström exponent $\mathring{A}_{par,bsc}(\lambda_1, \lambda_2)$ is needed. Furthermore, the particle depolarization ratio δ_{par} needs to be measured and the particle depolarization ratio of the background aerosol i.e. non-pollen particles δ_{bg} has to be known or reasonably well estimated. The method relies on the fact that pollen grains are coarse mode particles and presumably depolarizing (non-spherical shape), while background aerosol (e.g. pollution) consists of non-depolarizing, fine mode particles. The Ångström exponents and depolarization ratios of the two aerosol types (pollen and background aerosol) therefore differ. The total backscatter-related Ångström exponent is measured and the background depolarization ratio is determined as the mean value of pollen-free periods at the measurement site. To estimate the depolarization ratio of pure pollen, the Ångström exponent of pure pollen $\mathring{A}_{pollen}$ is needed. However, there are no such values in the literature. As pollen grains are quite big and thus can be assumed to be wavelength-independent on the backscatter at the lidar wavelengths 355 and 532 nm, we assume that the backscatter-related Ångström exponent at 355/532 nm should be 0. This assumption is necessary as otherwise it would not be possible to infer the depolarization ratio of pure pollen. However, in the future additional measurements to determine the Ångström exponent of pure pollen would be beneficial. The depolarization ratio of pure pollen δ_{pollen} can then be determined using an iterative approach. Table 5 summarizes the necessary parameters and the flow chart in Fig. 7 illustrates the method to retrieve the depolarization ratio values for pure pollen.

Using the measured total particle backscatter coefficient and depolarization ratio, the background depolarization ratio ($\delta_{bg} = 0.03$ in Vehmasmäki) and an estimated pollen depolarization ratio δ_x , it is possible to calculate the pollen backscatter $\beta_{pollen}(\delta_x, \lambda_2)$ following the method described by Tesche et al. (2009a). The pollen backscatter contribution $\chi_{pollen}(\delta_x, \lambda_2)$ is defined as the ratio of the

Table 5 Description of the parameters used to derive the pollen depolarization ratio.

measured	
β_{par}	total particle backscatter coefficient
δ_{par}	particle depolarization ratio
$\mathring{A}_{par,bsc}(\lambda_1, \lambda_2)$	backscatter-related Ångström exponent of total particles
δ_{bg}	background depolarization ratio (i.e. in the absence of pollen)
assumed	
$\mathring{A}_{pollen}$	backscatter-related Ångström exponent of pure pollen
δ_x	depolarization ratio of pure pollen; $\delta_x \in [\delta_{bg}, 1]$
calculated	
$\beta_{pollen}(\delta_x, \lambda_2)$	pollen backscatter coefficient; $\beta_{pollen} = \beta_{par} \frac{(\delta_{par} - \delta_{bg})(1 + \delta_{pollen})}{(\delta_{pollen} - \delta_{bg})(1 + \delta_{par})}$
$\chi_{pollen}(\delta_x, \lambda_2)$	pollen backscatter contribution; $\chi_{pollen}(\delta_x, \lambda_2) = \frac{\beta_{pollen}(\delta_x, \lambda_2)}{\beta_{par}(\lambda_2)}$
η	function of Ångström exponent; $\eta = \left(\frac{\lambda_1}{\lambda_2}\right)^{-\mathring{A}_{par}(\lambda_1, \lambda_2)}$

Figure 7 Flow chart of the retrieval method of the depolarization ratio value for pure pollen. Yellow boxes show measured (or simulated) parameters, blue boxes represent the assumptions/manual input and green boxes are the estimations/calculations. Figure adapted from Paper II © 2020 Atmospheric Chemistry and Physics.

pollen backscatter coefficient to the total particle backscatter coefficient and can theoretically range between 0, meaning no pollen, and 1 which means 100% of the aerosols are pollen. To investigate the relationship of the measured Ångström exponent $\mathring{A}_{par,bsc}(\lambda_1, \lambda_2)$ and the pollen backscatter contribution $\chi_{pollen}(\delta_x, \lambda_2)$, another parameter η is introduced which is a function of the Ångström exponent. For different assumed pollen depolarization ratios and therefore different pollen backscatter contributions, η gives a perfect linear relationship with χ . For each assumed δ_x , the η value for 100% pollen contribution ($\chi_{pollen}(\delta_x, \lambda_2) = 1$) can be defined as the value for pure pollen ($\eta_{pure}(\delta_x, \lambda_2)$). Assuming pure pollen has a backscatter-related Ångström exponent of 0, η for pure pollen should be 1. By iterating through different δ_x , the δ_x resulting in $\eta_{pure}(\delta_x, \lambda_2) = 1$ can be found. Finally, the δ_x when $\eta_{pure} = 1$ is the pollen depolarization ratio under the assumption that the Ångström exponent of pure pollen is zero.

This new method was applied to the lidar observations of the 2016 pollen campaign in Vehmasmäki to determine the depolarization ratio of pure pollen. Four intense pollination periods (IPPs) were chosen considering the pollen seasons, daily mean pollen concentration values of the dominant pollen type and the availability of lidar data. IPP-1 and IPP-2 are within the birch pollen season. During IPP-1 almost exclusively birch pollen was detected, while during IPP-2 a significant amount of spruce pollen was present. IPP-3 consists of two periods within the pine pollen season and IPP-4 was during the nettle pollen season. The optical properties of observed pollen layers in those periods were determined. As in **Paper I**, a wide range of lidar ratios and high backscatter-related Ångström exponents were observed. This indicates the dominance of fine-mode background aerosol to the backscatter signal and suggests that the lidar ratio and Ångström exponent alone are not suitable parameters to characterize pollen types. The depolarization ratio, however, showed enhanced values in the presence of pollen and even higher values in the presence of non-spherical pine and spruce pollen. The algorithm was then applied to the profiles detected during the IPPs assuming that there are only pollen and non-depolarizing background aerosols in the air. Under the assumption that the backscatter-related Ångström exponent of pure pollen between 355 and 532 nm is 0, a depolarization ratio of 0.24 at 532 nm and 0.17 at 355 nm for the birch period (IPP-1) was found. For the pine period (IPP-3) depolarization ratios of 0.36 at 532 nm and 0.30 at 355 nm were estimated (Table 6). These results suggest a spectral dependence of the depolarization ratio of pollen with higher depolarization ratio values at larger wavelengths. This could be a key factor for the detection and characterization of pollen and will be further investigated in **Paper IV**. Depolarization ratios for pure spruce and pure nettle pollen could not be retrieved with the obtained dataset. During IPP-2, spruce

pollen was always observed simultaneously with birch and with a varying mixing rate. During IPP-4 relatively small depolarization ratios with small variation have been observed for nettle pollen which complicated the separation.

Table 6 Linear depolarization ratio of birch and pine pollen under the assumption that the backscatter-related Ångström exponent between 355 and 532 nm for pollen is 0.

Pollen type	δ 532 nm	δ 355 nm
Birch	0.24 ± 0.01	0.17
Pine	0.36 ± 0.01	0.30

This study presents a method to determine the depolarization ratio of pollen when there is a mixture of a depolarizing (pollen) and a non-depolarizing aerosol type under the condition that the depolarization ratio of pollen is the only unknown variable. The depolarization ratio of the non-depolarizing aerosols needs to be known or reasonably well estimated. Furthermore, the Ångström exponent of total particles and pollen has to be known or estimated. As the Ångström exponent of pure pollen is not known so far, the assumption that it is 0 may cause a non-negligible bias. Thus, measuring the Ångström exponent of pure pollen, for example in laboratory experiments, would be beneficial and would certainly improve the determination of pure pollen depolarization ratios.

4.3 Utilization of Halo Doppler lidar depolarization to detect pollen

Currently, depolarization ratios are usually determined at the relatively short lidar wavelengths 355 and 532 nm and less often at the longer wavelengths 710 and 1064 nm. However, for certain studies, especially when investigating the spectral dependency, the depolarization ratio at longer wavelengths can provide additional information. In **Paper III**, we investigated the potential of commercially available Halo StreamLine Doppler lidars to obtain the depolarization ratio in the near infrared, at 1565 nm, to potentially extend the pollen studies with another wavelength. Halo StreamLine Doppler lidars are widely used for wind and turbulence studies, but using the post-processing recently developed by Vakkari et al. (2019), significantly weaker signals than before can be used. This opens up new possibilities for determining the depolarization ratio using Halo Doppler lidar measurements.

Figure 8 Averaged profiles on 15 May 2016, 19:00-21:00 UTC at Vehmasmäki. (a) Backscatter coefficient at 355, 532 and 1064 nm by Polly^{XT} and attenuated backscatter at 1565 nm by Halo Doppler lidar. (b) Particle linear depolarization ratio at 532 nm (Polly^{XT}) and 1565 nm (Halo). Error bars on the Halo depolarization profile represent measurement uncertainty. Figure from Paper III © 2021 Atmospheric Chemistry and Physics.

Halo StreamLine Doppler lidars can be equipped with a cross-polar receiver channel and co- and cross-polar signals can be detected consecutively. The depolarization ratio is calculated as the ratio of cross-polar to co-polar SNR, but in order to be used for aerosol characterization it needs to be calibrated. As Halo StreamLine Doppler lidars do not include a calibrator for the depolarization channel, the calibration needs to be done using the depolarization ratio at liquid cloud base. Spherical cloud droplets do not polarize the backscattered light and non-zero depolarization at the base of liquid clouds indicates incomplete extinction, or bleed-through, in the internal polarizer. After accounting for the bleed-through, the depolarization ratio at 1565 nm can be determined. Because of the long wavelength and low pulse energy, no molecular scattering is observed in the signal and the measured depolarization ratio can be considered as particle depolarization ratio. We compared the depolarization measurements of Halo Doppler lidars with co-

located measurements of two Polly^{XT} systems in different measurement environments. Dust, marine and polluted cases have been observed during a campaign from 21 April to 22 May 2017 in Limassol, Cyprus and atmospheric pollen, mainly birch and spruce pollen, has been observed from 9 to 16 May 2016 in Vehmasmäki, Finland. Figure 8 shows an example of backscatter and depolarization ratio profiles in Vehmasmäki during a day with a mixture of birch and spruce pollen. The particle depolarization ratio at 1565 nm is almost constant, while it decreases with height at 532 nm. This is probably caused by the higher sensitivity of the longer Halo wavelength to larger particles. The Halo signal is likely dominated by pollen grains, while at the shorter wavelength the increasing fraction of non-pollen particles with increasing height contributes to the signal. Comparisons under different aerosol conditions with the Polly^{XT} operated in Limassol, showed good agreement to previous studies at multiple wavelengths. For example for a case with fresh mineral dust, particle depolarization ratios at 1565 nm ranging from 0.29 to 0.32 were found and very low PDRs were found for polluted marine aerosol conditions.

Overall, Halo Doppler lidars can provide reasonable depolarization measurements with comparable quality to Polly^{XT} measurements when the aerosol signal is strong. Halo Doppler lidars have a much less powerful laser than Polly^{XT} systems, therefore the vertical extent of usable signal is limited. In the lowest 2-3 km, however, it can be used to add another wavelength to aerosol typing studies and can improve the distinction of different aerosol types using the spectral dependence.

4.4 Depolarization ratios at multiple wavelengths to characterize pollen

In **Paper I**, we have seen that the depolarization ratio can be used to detect pollen in the atmosphere. **Paper II** suggested a wavelength dependence of the depolarization ratio and **Paper III** showed that Halo Doppler lidars can be used to investigate the depolarization ratio at 1565 nm. Hence in **Paper IV**, we utilized the measurements of a co-located Halo Doppler lidar together with an upgraded Polly^{XT} lidar system during the pollen season 2019 in Vehmasmäki. The upgraded system has two depolarization channels at 355 and 532 nm and near-range capacities. Thus, it is possible to obtain profiles much closer to the ground (i.e. pollen source) and depolarization ratios at three wavelengths, 355, 532 and 1565 nm, can be detected.

Several periods with high pollen concentration have been observed. This study focuses on an intense birch and spruce pollen event from 16 - 19 May 2019 with

unusually high depolarization ratios. During this event maximal bi-hourly pollen number concentrations of up to 9000 grains per m^3 were observed. The amount of spruce pollen increased towards the end and a clearly different depolarization ratio was observed on the day with highest spruce concentration. Figure 9 shows a case study from each day of the 4-day period. The particle depolarization ratio at 532 and 1565 nm is clearly higher on the day with the highest spruce pollen share (see bar chart), while the PDR at 355 nm is around the same on all days. The increased depolarization on 19 May is likely linked to the large amount of spruce pollen. The PDR at 1565 nm is almost constant with height, while the depolarization at smaller wavelengths decreases with height. This behaviour has already been observed in **Paper III** and can be explained with the higher sensitivity of larger wavelengths to bigger particles such as pollen and lower sensitivity to smaller background aerosol. The depolarization ratios presented in this study are very high compared to commonly observed depolarization ratios for non-spherical particles. However, very high depolarization ratios of 0.5-1 have been observed for large dust, silt-sized and sand particles ($> 10 \mu\text{m}$) by Ansmann et al. (2009) and could also be characteristic for pollen grains considering their large size. Other possible reasons for such high depolarization ratios could be highly non-spherical pollen fragments caused by the rupture of bigger pollen grains or uneven surface

Figure 9 Case studies from each day of the 4-day intense period. Backscatter coefficient and particle depolarization ratio at 355 and 532 nm by Polly^{XT} (a-d), attenuated backscatter and particle depolarization ratio at 1565 nm by Halo Doppler lidar (e-f). The pollen concentration during the 2-h period from 9-11 UTC each day is shown in the bar chart. Green represents birch, red is representing spruce pollen. Figure from Paper IV © 2021 Atmospheric Chemistry and Physics.

structures of specific pollen types.

The whole period has been analyzed in 2-hour time intervals and investigated according to the share of spruce pollen. On the day with the highest spruce pollen share (22% of the total pollen number concentration), mean particle depolarization ratios of 0.46 ± 0.26 and 0.30 ± 0.09 were detected at 532 and 1565 nm respectively, in pollen layers down to around 350 m. Profiles at 355 nm could only be detected down to 900 m due to an overlap issue on the 355 nm far-range channel. Mean PDR values in pollen layers down to 900 m were 0.10 ± 0.02 , 0.38 ± 0.23 and 0.29 ± 0.10 at 355, 532 and 1565 nm, respectively. On the other days with significantly less spruce pollen contribution, the spectral dependence is much less pronounced (Fig. 10). Similar wavelength dependence with the maximum depolarization at the intermediate wavelength has already been observed in laboratory experiments for pine pollen by Cao et al. (2010). A wavelength dependence of the depolarization ratio might be characteristic for large, non-spherical pollen such as conifer pollen types and could be the key for pollen classification in automatic aerosol typing algorithms as it differs from other depolarizing aerosols like dust. Moreover, cases with a high share of spruce pollen were found to have a negative backscatter-related Ångström exponent. The Klett-Fernald method was used

Figure 10 Spectral dependence of the mean particle depolarization ratio of the days 16 - 19 May 2019. Laboratory-derived depolarization ratios by Cao et al. (2010) for pine and birch pollen are presented with empty markers. Error bars represent the standard deviation. Figure from Paper IV © 2021 Atmospheric Chemistry and Physics.

to derive the backscatter coefficients, the Ångström exponent is therefore directly affected by the assumption of the lidar ratio. However, different lidar ratios within reasonable range (30-80 sr) still lead to negative values. Negative Ångström exponents have previously been reported for mineral dust, but could also be a characteristic feature of spruce pollen. Those features can help to distinguish pollen from other aerosol types and improve aerosol typing algorithms. To determine, however, whether whole non-spherical pollen grains are causing the high depolarization ratios or pollen fragments and the pollen surface are contributing to the observations as well, more studies are needed. Non-spherical pollen fragments and the surface structure of specific pollen types could possibly affect the observed depolarization ratio, but could not be determined in this study. In future studies, the amount of pollen fragments should be recorded and the effect of surface patterns needs to be determined. This could be achieved using ground-based or airborne in-situ measurements for example onboard balloons or drones.

5 Conclusions

Atmospheric pollen is not only the cause of many allergy-related diseases, it also affects the climate by directly scattering sunlight and by acting as CCN and INP and therefore affecting cloud formation. Thus, a good understanding of the distribution and transport of pollen is needed to verify and improve pollen and climate models. However, pollen type and concentration are usually measured on ground level and information about the vertical distribution is mostly missing. Lidar is an excellent tool to investigate the vertical distribution of aerosols in the atmosphere as measurements from ground to several kilometers altitude are possible with high vertical and temporal resolution and without affecting the aerosol in its ambient conditions.

In this study, we investigated how the lidar technique can be used to detect and characterize pollen in the atmosphere. The first objective of this thesis, stated in the introduction, was to explore the capabilities of multiwavelength Raman polarization lidars for the detection and characterization of pollen. We found that in the presence of pollen, especially non-spherical spruce pollen, enhanced depolarization ratios can be observed (**Paper I**). The depolarization ratio seems to be the most useful parameter for the characterization of different pollen types when they exhibit different shapes. The presented observations suggest that the backscattering, lidar ratio and Ångström exponent may be dominated by background aerosol and should be considered with caution for pollen characterization. **Paper I** has demonstrated that the detection of pollen, although challenging, is possible. It has also been shown that certain points need to be addressed in order to improve the detection of pollen. These are for example the minimal detection height, which has been addressed in **Paper IV** by adding a near-field unit to the operated lidar system and the importance of the depolarization ratio which is the main focus of **Paper II to IV**.

The second objective was to investigate the depolarization ratio of pure pollen. Using lidar measurements we always observe a mixture of different aerosols in the atmosphere, but to characterize the optical properties of pollen, they need to be separated from the background aerosol. In **Paper II** we presented a method to estimate the depolarization ratio of pollen using lidar measurements. The method uses the assumption that the backscattering on pure pollen is wavelength independent and the backscatter-related Ångström exponent is therefore zero. Under this assumption, a depolarization ratio of 0.24 and 0.36 at 532 nm and 0.17 and 0.30 at 355 nm has been found for birch and pine, respectively.

In **Paper III**, we explored if Halo Doppler lidar measurements can be used to add another depolarization wavelength to our pollen studies. In the presented study, we showed that after applying a calibration method using the depolarization ratio

at liquid cloud base, Halo Doppler lidars can provide reasonable depolarization measurements at least in the lowest 2-3 km. The detected depolarization ratio can be considered as particle depolarization ratio because of the long wavelength and low pulse energy of Halo Doppler lidars. A comparison with co-located Polly^{XT} measurements during the campaign in Vehmasmäki 2016 showed an almost constant PDR at 1565 nm in contrast to the PDR at 532 nm for cases with birch and spruce pollen in the atmosphere. This is an interesting finding and could be explained by the higher sensitivity of longer wavelengths to larger particles such as pollen.

As result of the previous studies, we utilized measurements of an upgraded version of Polly^{XT} with near-range capacities and a second depolarization channel and co-located Halo Doppler lidar measurements during a second pollen campaign in Vehmasmäki. In **Paper IV**, depolarization ratios at three wavelengths 355, 532 and 1565 nm are presented and it is investigated if the depolarization ratio at multiple wavelengths can be used to characterize atmospheric pollen. We found that in the presence of large, non-spherical spruce pollen, depolarization ratios at 532 and 1565 nm are clearly higher than on days with less or no spruce pollen contribution. There is a wavelength dependence of the depolarization ratio with highest value at 532 nm, which could be characteristic for large, non-spherical pollen such as conifer pollen. This wavelength dependence could be the key for recognizing and classifying pollen as it differs pollen from other depolarizing aerosol.

Overall, the presented results demonstrate that multiwavelength lidar measurements can be used to track the presence of pollen in the atmosphere. Especially the depolarization ratio seems to be a valuable parameter for the detection and characterization of pollen. For the first time, depolarization ratios of atmospheric pollen in ambient conditions were detected at three wavelengths. Depolarization measurements obtained with Halo Doppler lidar proved to be a useful addition to measurements with a Polly^{XT} Raman lidar. A spectral dependence of the depolarization ratio has been found which could be an indicator for non-spherical pollen. Nevertheless, future studies are needed to obtain a more comprehensive database. Especially observations with only one pollen type are needed to determine the optical properties of a specific pollen type in the atmosphere. These values are needed as otherwise the observed values are only representative for the exact same mixture of the observed pollen types. Also determining the Ångström exponent of pure pollen, for example in laboratory experiments, would improve the estimation of the depolarization ratio of pure pollen and therefore the detection and the classification of pollen using depolarization ratio values. In future studies, pollen fragments should be recorded to identify the impact of whole pollen grains and smaller pollen fragments to the lidar measurements. Drone- or balloon-based

in-situ observations could be used to complement the lidar observations and to determine the pollen mixing with height. Furthermore, the application of other lidar systems e.g. fluorescence lidars in combination with Raman polarization lidar measurements could significantly improve the detection of pollen. Moreover, ways to separate different depolarizing aerosol types need to be explored, as otherwise it will be difficult to implement the detection of pollen using polarization lidar measurement in regions other than remote forest areas.

The results of this thesis present a valuable basis for future studies and can help to improve the detection and classification of pollen. Correctly classified pollen could enable an almost near-real time detection of pollen, not only on ground level but also in the air. Applying pollen detection to satellite based aerosol classifications, for example to Calipso or EarthCare classifications, would furthermore extend pollen detection to global scale. Lidar profiles could then be used to evaluate pollen dispersion models and subsequently improve pollen forecasts which would substantially improve the well-being of allergy sufferers.

6 Author's contribution

- Paper I** Bohlmann, S., X. Shang, E. Giannakaki, M. Filioglou, A. Saarto, S. Romakkaniemi, and M. Komppula (2019), Detection and characterization of birch pollen in the atmosphere using a multiwavelength Raman polarization lidar and Hirst-type pollen sampler in Finland, *Atmospheric Chemistry and Physics*, 19(23), 14559–14569, doi: 10.5194/acp-19-14559-2019
Author's contribution: The author was responsible for conceptualizing the methodology with support of the supervisors. Furthermore, the author analyzed the lidar data and wrote the manuscript.
- Paper II** Shang, X., E. Giannakaki, S. Bohlmann, M. Filioglou, A. Saarto, A. Ruuskanen, A. Leskinen, S. Romakkaniemi, and M. Komppula (2020), Optical characterization of pure pollen types using a multiwavelength Raman polarization lidar, *Atmospheric Chemistry and Physics*, 20(23), 15323–15339, doi: 10.5194/acp-20-15323-2020
Author's contribution: The author contributed to the conceptualizing of the methodology and contributed to the scientific discussion.
- Paper III** Vakkari, V., H. Baars, S. Bohlmann, J. Bühl, M. Komppula, R.-E. Mamouri, and E. J. O'Connor (2021), Aerosol particle depolarization ratio at 1565 nm measured with a Halo Doppler lidar, *Atmospheric Chemistry and Physics*, 21(8), 5807–5820, doi: 10.5194/acp-21-5807-2021
Author's contribution: The author performed the data analysis of the Raman lidar data and contributed to the scientific discussion.
- Paper IV** Bohlmann, S., X. Shang, V. Vakkari, E. Giannakaki, A. Leskinen, K. E. J. Lehtinen, S. Pätsi, and M. Komppula (2021), Lidar depolarization ratio of atmospheric pollen at multiple wavelengths, *Atmospheric Chemistry and Physics*, 21(9), 7083–7097, doi: 10.5194/acp-21-7083-2021
Author's contribution: The author was responsible for the lidar observations and the collection of the Burkard pollen samples. Furthermore, the author performed the data analysis and conceptualized and wrote the manuscript with support of the co-authors.

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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 Polly^{XT} 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-

tribution, however, is missing. Models are using phenological and meteorological data to forecast concentration and dispersion of pollen in the atmosphere. But observations, especially in the vertical direction, to evaluate the model 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. Recently, the signature of pollen grains has been observed in lidar measurements (Noh et al., 2013a, b; Sassen, 2008; Sicard et al., 2016). It has been revealed that nonspherical pollen generates strong laser depolarization, and thus information on particle shape can be retrieved. For example, Sassen (2008) measured a linear depolarization ratio at 694 nm up to 30 % for paper birch in Alaska. In controlled laboratory experiments, Cao et al. (2010) measured the linear depolarization ratio of different pollen types and determined a linear depolarization ratio at 532 nm for paper birch of 33 %. In the absence of other depolarizing particles, the depolarization ratio could therefore be used to track pollen grains. Lidar-derived depolarization ratio profiles thus can provide information about the vertical distribution of pollen, which could result in an improvement in the model input parameters and serve as validation for model results.

Within our study, we aim to improve and deepen the knowledge of optical properties of pollen in the atmosphere by using a multiwavelength Raman polarization lidar. Finland provides suitable conditions for the observation of pollen as 78 % of Finland's total area is forestland and sources of other highly depolarizing particles like dust are absent. Hence the contamination with other aerosols is considered to be small. During a 4-month measurement campaign in 2016, the multiwavelength Raman polarization lidar Polly^{XT} (Engelmann et al., 2016) 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 birch and spruce pollen as birch is one of the most allergenic pollen types and the most allergenic tree pollen in northern, central and eastern Europe (D'Amato et al., 2007).

2 Measurement site and instrumentation

Our measurement campaign took place from the beginning of May to the end of August 2016 at Vehmasmäki, Finland (62°44' N, 27°33' E; 190 m a.s.l.); a rural forest site 18 km from the city center of Kuopio, Eastern Finland. The measurement site was equipped with the multiwavelength Raman polarization lidar Polly^{XT} (Sect. 2.1) and a Hirst-type pollen sampler (Sect. 2.2). With this setup it is possible to combine

vertical profiles of the aerosol properties above the site and the pollen concentration on the ground. Due to the location of the site, far from major aerosol sources like dust or anthropogenic aerosol and mainly surrounded by forest, the atmosphere is relatively clean and suitable for pollen studies.

2.1 Lidar: Polly^{XT}

Lidar measurements were conducted with the multiwavelength Raman polarization lidar Polly^{XT} (Althausen et al., 2009; Baars et al., 2016; Engelmann et al., 2016). Polly^{XT} 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 during dark hours (Filioglou et al., 2017). The simultaneous measurement of the cross-polarized and total backscattered light at 532 nm enables the determination 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 ($\text{\AA}$), 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 about 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

uncertainties propagate to the retrieved Ångström exponents and LR_s.

Further details on the setup, principle and error propagation of Polly^{XT} can be found in Althausen et al. (2009) and Engelmann et al. (2016). Near-real-time measurements and Polly^{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⁻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⁻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⁻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-Ščevková, 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.

Figure 1. Overview of the 11 d birch pollen period. (a) Pollen concentration obtained from the Hirst-type pollen sampler, (b) range-corrected signal at 1064 nm and (c) volume depolarization ratio at 532 nm. Dashed vertical lines mark the period of the case studies.

Figure 2. Layer definition during the measurement period 5–15 May 2016. The definition of layers is explained in Sect. 3. Colors mark the upper layers, which were not used for further analysis. Triangles mark the lower and upper limits of the area used for calculation.

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 of 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. (a, c) 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 , respec-

tively. 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,

Figure 4. Pollen micrographs are shown. **(a)** Birch (*Betula pendula*) pollen grain (source: Halbritter and Diethart, 2016). **(b)** Spruce (*Picea abies*) pollen grains (source: the Biodiversity Unit of the University of Turku, Kari Kaunisto).

respectively. The backscatter-related Ångström exponent at 532–1064 nm is 0.8 to 1.0.

The PDR at 532 nm is on average $19 \pm 2\%$, 24 % at its maximum, and is clearly higher than in the case when only birch pollen was observed (period 1). The air masses arriving at the height of the layers on 15 May 20:00 UTC have been advected from Russia and remained close to the ground only for the last 12 h before reaching the site. The contamination with depolarizing dust can therefore be neglected. An explanation for the higher depolarization of the backscattered light is the nonspherical shape of the spruce pollen grains which have been detected in addition to birch pollen.

Figure 4 shows micrographs of birch and spruce pollen grains. Birch pollen (Fig. 4a) has a diameter around 20–30 μm , is almost spherical and possess three pores on the edge of the grain. Spruce pollen grains, on the other hand, possess two air bladders and are clearly nonspherical. Furthermore, those pollen grains are significantly larger, with their longest-axis diameter ranging between 90 and 110 μm (including air bladders).

Pollen grains are low-density particles, which make them more sensitive to air currents, reduces the settling velocity and allows them to be lifted by turbulent air flows. Birch pollen, for example, has 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 μm (PM_{10}) 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 has been shown by several studies on the long-distance transport of pollen (Rousseau et al., 2008; Skjth et al., 2007; Szczepanek et al., 2017).

4.2 Lidar-derived optical parameters

All pollen layers between 5 and 15 May have been identified and analyzed to determine the relationship between pollen

type and the lidar-derived optical properties of the aerosol layer. Figure 5a shows the LR at 532 nm against the PDR at the same wavelength for all Raman measurements during nighttime. Measurements during the first intense birch pollination period are shown in green, while measurements during the second period, when spruce pollen was detected simultaneous to birch pollen, are shown in black. The size of the dots represents the measured pollen concentration by the Hirst-type sampler at ground level. The standard deviation is shown by the error bars. Lidar ratio values range from 31 to 74 sr. This wide range of LRs suggest that the LR alone is not a suitable parameter for the characterization of pollen as other aerosol types also show characteristic values in this range. However, the mean PDR within the pollen layers during the first period is $10 \pm 6\%$, which is significantly higher than that of anthropogenic pollution. In the absence of other depolarizing aerosol, e.g., dust, pollen is likely the dominant aerosol causing this depolarization. Depolarization ratios higher than 15 % are only observed during the second period (12–15 May) in which spruce pollen was present. The mean LR and PDR at 532 nm during this period are $62 \pm 10 \text{ sr}$ and $21 \pm 3\%$, respectively. The significantly higher PDR is caused by the nonspherical shape of spruce pollen in those layers. In Fig. 5b, the backscatter-related Ångstrm exponent between 532 and 1064 nm is shown against the PDR at 532 nm for all measurements between 5 and 15 May, including Klett retrievals for daytime measurements (dotted markers). A clear tendency towards smaller Ångstrm exponents with increasing depolarization ratios in both periods can be seen. This correlation indicates that the influence of nonspherical particles on the backscattered signal increases with decreasing Ångstrm exponent, i.e., bigger particles. The Ångstrm 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 ($\sim 90\text{--}110 \mu\text{m}$), even with a small contribution ($\sim 14\%$) to the total pollen number concentration. But considering the different volumes of birch and spruce pollen grains, the contribution of spruce to the total volume concentration exceeds 75 % in the second period, which may explain the large effect of spruce pollen on the measured optical properties even with a small number concentration.

However, the effect of the background particles has to be considered. Lidar measurements during the winter months of 2015 and 2016 and during pollen-free periods in spring and summer 2016 have been analyzed to determine the effect of background aerosol at our measurement site. During winter time the absence of pollen can be ensured but there is a possibility that pollen also had been present in the atmosphere during spring and summer when no pollen was detected by the Hirst-type sampler on the ground. Nevertheless, values of mean PDR at 532 nm are below 4 % during all analyzed periods with no observed pollen. Since the PDR during the pollination period is significantly higher than the PDR of the background aerosol, the depolarization ratio can

Figure 5. Mean values of optical properties of the detected pollen layers during the period 5 to 15 May. Error bars denote the standard deviation, and marker size denotes the pollen concentration. Marker color denotes the measurement period. Green dots are measured from 5 to 10 May and black dots from 12 to 15 May. Solid dots indicate Raman retrievals and dotted markers the Klett solution. Panel (a) shows lidar ratio at 532 nm vs. particle depolarization ratio at 532 nm. Panel (b) shows backscatter-related Ångström exponent at 532–1064 nm vs. particle depolarization ratio at 532 nm.

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

Figure 6. Dependence of the backscatter-related Ångström exponent at 532–1064 nm (a) and the particle depolarization ratio at 532 nm (b) on the relative humidity for all Raman measurements of pure and mixed birch cases during the first period (5–10 May, green) and the second period (12–15 May, black).

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 $\pm$ SD	Range	Median
Layer top height (km)		1.3 ± 0.3	1.0–2.2	1.2
Backscatter coefficient ($\text{Mm}^{-1} \text{sr}^{-1}$)	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 $\pm$ SD	Range	Median
Layer top height (km)		1.3 ± 0.4	1–2.2	1.1
Backscatter coefficient ($\text{Mm}^{-1} \text{sr}^{-1}$)	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 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 (Teschke et al., 2011) or dust mixtures with marine aerosol (Groß et al., 2011), thus pollen could easily be misclassified as dusty mixtures. The mean LR_s show a wide range of values depending on the mixing of differ-

ent 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 LR_s are characteristic for dust or dust–smoke mixtures (Teschke 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).

Author contributions. SB, XS and MF performed the lidar data analysis. AS analyzed the pollen samples. MK and EG initiated the measurement campaign. All authors contributed to the scientific discussion and the article preparation.

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

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Abstract. We present a novel algorithm for characterizing the optical properties of pure pollen particles, based on the depolarization ratio values obtained in lidar measurements. The algorithm was first tested and validated through a simulator and then applied to the lidar observations during a 4-month pollen campaign from May to August 2016 at the European Aerosol Research Lidar Network (EARLINET) station in Kuopio (62°44' N, 27°33' E), in Eastern Finland. With a Burkard sampler, 20 types of pollen were observed and identified from concurrent measurements, with birch (*Betula*), pine (*Pinus*), spruce (*Picea*), and nettle (*Urtica*) pollen being the most abundant, contributing more than 90 % of the total pollen load, regarding number concentrations. Mean values of lidar-derived optical properties in the pollen layer were retrieved for four intense pollination periods (IPPs). Lidar ratios at both 355 and 532 nm ranged from 55 to 70 sr for all pollen types, without significant wavelength dependence. An enhanced depolarization ratio was found when there were pollen grains in the atmosphere, and an even higher depolarization ratio (with mean values of 0.25 or 0.14) was observed with the presence of the more non-spherical spruce or pine pollen. Under the assumption that the backscatter-related Ångström exponent between 355 and 532 nm should be zero for pure pollen, the depolarization ratio of pure pollen particles at 532 nm was assessed, resulting in 0.24 ± 0.01 and 0.36 ± 0.01 for birch and pine pollen, respectively. Pollen optical properties at 1064 and 355 nm were also estimated. The backscatter-related Ångström exponent between 532 and 1064 nm was assessed to be ~ 0.8 (~ 0.5) for pure birch

(pine) pollen; thus the longer wavelength would be a better choice to trace pollen in the air. Pollen depolarization ratios of 0.17 and 0.30 at 355 nm were found for birch and pine pollen, respectively. The depolarization values show a wavelength dependence for pollen. This can be the key parameter for pollen detection and characterization.

1 Introduction

Pollen has various effects on human health and the environment. The number of people suffering from allergies due to pollen inhalation is rising (Schmidt, 2016). Airborne pollen is recognized as one of the major agents of allergy-related diseases such as asthma, rhinitis, and atopic eczema (Bousquet et al., 2008). Pollen is also a biogenic air pollutant which affects both the solar radiation reaching the Earth and cloud optical properties by acting as nuclei for both cloud droplets and ice crystals (Steiner et al., 2015).

Various networks are built to monitor pollen concentrations at ground level using in situ instruments (Giesecke et al., 2010). In 2020, there are more than 1000 active pollen monitoring stations in the world (Buters et al., 2018; https://oteros.shinyapps.io/pollen_map/, last access: 7 April 2020), with the majority based on the Hirst principle (Hirst, 1952). The conventional method of pollen classification is based on pollen morphological characteristics using microscopy (Holt and Bennett, 2014; Weber, 1998). However, it requires complex procedures for complete classification and identifica-

tion, and the results are not publicly available online. Besides, pollen grains can be agile and change their visual nature before the analysis, e.g. undergo an osmotic shock (Miguel et al., 2006), which leads to errors in pollen characterization. Several studies on the long-distance transport of pollen (Rousseau et al., 2008; Skjth et al., 2007; Szczepanek et al., 2017) have shown that pollen grains can be lifted up to several kilometres and be dispersed by wind over thousands of kilometres.

An increasing interest in pollen has arisen in the aerosol lidar community (Noh et al., 2013; Sicard et al., 2016). In our previous study (Bohlmann et al., 2019) we showed on the basis of an 11 d birch pollination period that lidar measurements can detect the presence of pollen grains in the atmosphere and that non-spherical pollen grains can generate strong depolarization (we found a mean depolarization ratio of 0.26 for the birch–spruce pollen mixture). Therefore, it is possible to observe airborne pollen grains in the atmosphere using the depolarization ratio in the absence of other depolarizing non-spherical particles (e.g. dust). We have also reported that lidar-derived parameters (e.g. depolarization ratio and Ångstrm exponent) provide the possibility of identifying different pollen types (e.g. birch and spruce pollen). However, the optical properties of pure pollen are still missing due to the fact that the atmospheric aerosol population is always a mixture of several particle types. For instance, the depolarization ratio of pure pollen is an essential parameter needed to separate pollen backscatter from the background aerosol backscatter. The Ångstrm exponent and the lidar ratio, which are often used for aerosol typing, are also crucial parameters to be defined for pure pollen particles.

In this study, we present a novel method for characterizing the optical properties of pure pollen particles, based on a 4-month campaign. In Sect. 2, we introduce the pollen campaign and the instruments. In Sect. 3, we present the methodology and describe a novel algorithm to estimate the depolarization ratio value for pure pollen. This algorithm is tested and validated through a simulator. In Sect. 4, we report the results: firstly, the pollen information observed by the Burkard sampler and lidar-retrieved optical properties for the pollen layer are presented. Secondly, the novel algorithm of Sect. 3 is applied to the lidar observations in Sect. 4.3 to retrieve the optical properties for pure pollen. Section 5 is devoted to the summary and conclusion.

2 Site and instruments

The measurement campaign was performed from May to August 2016 at the Kuopio station of the European Aerosol Research Lidar Network (EARLINET) in Vehmasmki (6244' N, 2733' E; elevation of 190 m above sea level). This rural site is mainly surrounded by forest, located ~ 18 km from the city centre of Kuopio, in Eastern Finland. Finland provides suitable conditions for the observation of

pollen as 78 % of Finland's total area is covered by forests. Airborne *Betula* spp. (birch) pollen is one of the most recognized aeroallergens in northern European countries and the most important cause of pollen allergy (Sofiev et al., 2015; Yli-Panula et al., 2009). The predominant *Betula* species include *B. pendula* and *B. pubescens*, while *B. nana* and *B. pubescens* subsp. *czerepanovii* can be found in northern parts of the country. As for conifers, *Pinus sylvestris* and *Picea abies* are the most prevalent, and *P. sylvestris* pollen typically causes the highest peaks during the pollen season. *P. sylvestris* and *P. abies* are the only naturally growing species of their genera in Finland. Compared to many other European countries, relatively clean background atmospheric conditions in Finland favour pollen detection and further separation of contributions of pollen backscattering from total scattering by using lidars, since there are fewer other particles, particularly dust, which would complicate the analysis.

The Kuopio station is operated by the Finnish Meteorological Institute, and it has been equipped with a ground-based multi-wavelength Raman polarization lidar Polly^{XT} (Engelmann et al., 2016), Doppler lidar, and in situ instruments next to a 318 m tall mast (for the meteorological observations) since autumn 2012 (Hirsikko et al., 2014). Polly^{XT} has three emission wavelengths (355, 532, and 1064 nm) and seven detection channels (including three emitted wavelength channels, three inelastic Raman-shifted wavelength channels (387, 407, and 607 nm), and the cross-polarization channel at 532 nm). Polly^{XT} has an initial spatial resolution of 30 m and a temporal resolution of 30 s. 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 a 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 the 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-polarization and total polarization channels of the Polly^{XT} allow the retrieval of the volume linear depolarization ratio (VDR) and particle linear depolarization ratio (PDR) at 532 nm, which provide information on the shape of the scattering particles. Multi-wavelength measurements (355, 532, and 1064 nm) enable the determination of Ångstrm exponents between each wavelength pair, which are related to the particle nature, mostly the size. Previous studies show (e.g. Eck et al., 1999) that Ångstrm exponent values greater than 2 indicate small particles associated with combustion byproducts, whereas Ångstrm exponent values less than 1 indicate large particles like sea salt and dust.

In addition to the lidar measurements, a Hirst-type Burkard pollen sampler (Hirst, 1952) was placed 4 m above ground level (a.g.l.) next to the lidar instrument. The Burkard sampler enables identification of pollen types and concentration microscopically with a 2 h time resolution. More detailed descriptions of the pollen sampler and Polly^{XT} used during this campaign can be found in Bohlmann et al. (2019) and reference therein.

3 Methodology – a synthetic simulator

In this study, we provide a novel method and develop an algorithm to estimate the depolarization ratio value for pure pollen particles. This algorithm is first tested through a simulator (Sect. 3) using the synthetic lidar data and then applied to the real lidar observations (Sect. 4.3). The simulator includes a direct model and an inverse model module (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. We mainly consider two wavelengths: $\lambda_1 = 355$ nm and $\lambda_2 = 532$ nm. Other wavelength combinations of 532 and 1064 nm will be briefly discussed at the end of Sect. 3.1.

3.1 Direct model – generation of synthetic optical profiles

Two aerosol populations, pollen (depolarizing) and background (non-depolarizing) aerosols, are considered in this simulation. The optical and physical parameters used in the direct calculation are presented in Table 1; these parameters are named “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 ratios of non-pollen particles ($\delta_{\text{background}}$) at both 355 and 532 nm are selected to be 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 ratios of pure pollen particles (δ_{pollen}) at 532 nm 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 ($\text{\AA}_{\text{pollen}}$) of 0. The backscatter-related Ångström exponent of non-pollen particles ($\text{\AA}_{\text{background}}$) between 355 and 532 nm 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 two constraints: (i) the depolarization ratio of pollen (depolar-

izing one) should be higher than the depolarization ratio of background aerosol (non-depolarizing one); and (ii) the values of backscatter-related Ångström exponent for pollen and non-pollen particles should be different. In addition, the conclusion of the simulation section is not dependent on the assumed profile shape or height; and the initial values are not critical for presenting the overall approach.

The extinction coefficient profiles of these two aerosol layers are assumed to follow a Gaussian distribution. The optical depth (OD) of the input background aerosol layer is fixed to be 0.1 in this simulation. In order to simulate different pollen contributions to the total aerosol load, we change the pollen load by selecting different input values for the pollen layer OD. A pollen OD of 0.002, 0.01, 0.02, 0.05, 0.1, and 1 is used; thus six pollen backscatter coefficient profiles are simulated. One example of simulated pollen and background backscatter coefficients is shown in the Supplement (Fig. S2a) for a pollen OD of 0.1. The pollen layer is defined as the layer below 1 km.

Next, the pollen layer and background layer are summed up (Eq. 1), and then the vertical profiles of aerosol backscatter coefficient, lidar ratio, and Ångström exponent of the total particles are simulated (e.g. Fig. S2b). The Ångström exponent describes the wavelength dependence on aerosol optical properties (Ångström, 1964). The backscatter-related Ångström exponent between two wavelengths of λ_1 and λ_2 (denoted as $\text{\AA}$) can be expressed as Eq. (2).

$$\beta_{\text{particle}}(\lambda, z) = \beta_{\text{pollen}}(\lambda, z) + \beta_{\text{background}}(\lambda, z) \quad (1)$$

$$\text{\AA}_x(\lambda_1, \lambda_2, z) = -\frac{\ln\left(\frac{\beta_x(\lambda_1, z)}{\beta_x(\lambda_2, z)}\right)}{\ln\left(\frac{\lambda_1}{\lambda_2}\right)} \quad (2)$$

The index $x = \text{pollen, background, or particle}$ denotes the backscatter-related Ångström exponent of pollen, background, or total particles.

Vertical profiles of the particle linear depolarization ratio (PDR; denoted as δ_{particle}) can be also calculated following Eq. (3) (the detailed calculation is given in the Supplement).

$$\delta_{\text{particle}} = \frac{\frac{\beta_{\text{pollen}} \cdot \delta_{\text{pollen}}}{\delta_{\text{pollen}} + 1} + \frac{\beta_{\text{background}} \cdot \delta_{\text{background}}}{\delta_{\text{background}} + 1}}{\frac{\beta_{\text{pollen}}}{\delta_{\text{pollen}} + 1} + \frac{\beta_{\text{background}}}{\delta_{\text{background}} + 1}} \quad (3)$$

Theoretically, these parameters can be derived directly from lidar observations. In order to keep the consistency of the availability of lidar-derived parameters, particle backscatter coefficients at 532 nm, PDRs at 532 nm, and backscatter-related Ångström exponents between 355 and 532 nm simulated for these six cases (shown in Fig. 1) will be used later as input for the inverse model.

The pollen backscatter contribution, denoted as χ_{pollen} (Eq. 4), is defined as the ratio of pollen backscatter coefficient (β_{pollen}) to the total particle backscatter coefficient (β_{particle}). Note that the use of “particle” here is to distinguish

Table 1. Parameters of pollen and background aerosol layers as input for the direct model. LR: lidar ratio, DR: depolarization ratio, $\text{\AA}$ bsc: backscatter-related Ångström exponent. A Gaussian distribution is applied for each layer, with the layer centre and half width given.

Aerosol type	LR 355 nm (sr)	LR 532 nm (sr)	DR (δ) 355 nm	DR (δ) 532 nm	$\text{\AA}$ bsc 355–532 nm	Layer centre	half width (Gauss.)
Pollen	65	65	0.35	0.35	0	0.5 km	1 km
Background	50	50	0.03	0.03	2	1.5 km	3 km

Figure 1. Six cases of simulated vertical profiles of (a) particle backscatter coefficient at 532 nm, (b) particle linear depolarization ratio at 532 nm, and (c) backscatter-related Ångström exponent between 355 and 532 nm. Simulated results under different input pollen optical depth (OD) values are shown by colour.

from “molecular”.

$$\chi_{\text{pollen}}(\lambda, z) = \frac{\beta_{\text{pollen}}(\lambda, z)}{\beta_{\text{particle}}(\lambda, z)} \quad (4)$$

We investigate here the relationship of the backscatter-related Ångström exponent of total particles ($\text{\AA}_{\text{particle}}$) and pollen backscatter contribution (χ_{pollen}) at different wavelengths (the detailed calculation is given in the Supplement), resulting in a power law relationship:

$$\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{\AA}_{\text{particle}}(\lambda_1, \lambda_2)} = \left(\left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{\AA}_{\text{pollen}}(\lambda_1, \lambda_2)} - \left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{\AA}_{\text{background}}(\lambda_1, \lambda_2)}\right) \cdot \chi_{\text{pollen}}(\lambda_2) + \left(\frac{\lambda_1}{\lambda_2}\right)^{-\text{\AA}_{\text{background}}(\lambda_1, \lambda_2)}. \quad (5)$$

The wavelength pairs (λ_1, λ_2) are selected as (355, 532), (532, 355), or (1064, 532) in this study. In order to simplify the calculation, we introduce two parameters, η and η' , as a function of the backscatter-related Ångström exponent between 355 and 532 nm or between 532 and 1064 nm, for the total particle backscatter coefficients:

$$\begin{cases} \eta = \left(\frac{355}{532}\right)^{-\text{\AA}_{\text{particle}}(355, 532)} \\ \eta' = \left(\frac{1064}{532}\right)^{-\text{\AA}_{\text{particle}}(1064, 532)} \end{cases} \quad (6)$$

The linear relationships resulting from the pairs of parameters η or η' and χ_{pollen} at different wavelengths are reported in Table 2. For example, the pollen backscatter contribution at 532 nm ($\chi_{\text{pollen}}(532)$) is inversely proportional to the parameter η . Using the previous six simulated cases, a perfect linear relationship is found to fit the η versus $\chi_{\text{pollen}}(532)$ (Fig. 2).

3.2 Inverse model – retrieval of depolarization ratio

In this section, we present the inverse model to retrieve the depolarization ratio of pure pollen particles. Tesche et al. (2009) provide a method to separate dust and non-dust contributions, based on the difference of the depolarization ratio values of these two types. This separation method is applied here to separate the two simulated aerosol types.

The pollen backscatter coefficient can be separated from the total particle backscatter coefficient (calculated from Eq. 3), expressed as

$$\beta_{\text{pollen}} = \beta_{\text{particle}} \frac{(\delta_{\text{particle}} - \delta_{\text{background}})(1 + \delta_{\text{pollen}})}{(\delta_{\text{pollen}} - \delta_{\text{background}})(1 + \delta_{\text{particle}})}. \quad (7)$$

The only remaining unknown to solve the Eq. (7) is the depolarization ratio for pure pollen (δ_{pollen}). Next we use previously simulated β_{particle} and δ_{particle} and the assumed $\delta_{\text{background}}$. From now on, 532 nm will be the default wavelength (if not otherwise specified). The wavelength pair

Table 2. The linear relationships resulting from the pairs of parameters $\eta(\hat{A}_{\text{particle}})$ and χ_{pollen} at different wavelengths are reported.

Wavelength pair (λ_1, λ_2) (nm)	Pollen backscatter contribution at λ_2	Backscatter-related Ångström exponent $\hat{A}(\lambda_1, \lambda_2)$	Parameter of $\hat{A}(\lambda_1, \lambda_2)$ linearly correlating with χ_{pollen}	Formulae
$\lambda_1 = 355, \lambda_2 = 532$	$\chi_{\text{pollen}}(532)$	$\hat{A}_{\text{particle}}(355, 532)$	η	$\eta = \left(\frac{355}{532}\right)^{-\hat{A}_{\text{particle}}(355, 532)}$
$\lambda_1 = 532, \lambda_2 = 355$	$\chi_{\text{pollen}}(355)$	$\hat{A}_{\text{particle}}(532, 355)$	$\frac{1}{\eta}$	$\frac{1}{\eta} = \left(\frac{532}{355}\right)^{-\hat{A}_{\text{particle}}(355, 532)}$
$\lambda_1 = 1064, \lambda_2 = 532$	$\chi_{\text{pollen}}(532)$	$\hat{A}_{\text{particle}}(1064, 532)$	η'	$\eta' = \left(\frac{1064}{532}\right)^{-\hat{A}_{\text{particle}}(532, 1064)}$

Figure 2. Scatter plot using the parameter $\eta = (355/532)^{-\hat{A}(355, 532)}$ and pollen backscatter contribution (χ_{pollen}) at 532 nm for six simulated cases, of which the input values of pollen optical depth ($\text{OD}_{\text{pollen}}$) at 532 nm are defined as 0.002, 0.01, 0.02, 0.05, 0.1, and 1 (shown on the bottom x axis), and the input value of background optical depth is fixed to be 0.1. Mean values of pollen layers (0–1 km) are used for χ_{pollen} and η . They line up perfectly following Eq. (5).

(λ_1, λ_2) is selected as (355, 532) in this section. Mean values of optical properties inside the pollen layer are considered in this study; it is also possible to use values of each bin of the synthetic profile which will lead to the same conclusion. Mean values of backscatter-related Ångström exponent between 355 and 532 nm inside the pollen layer, denoted as $\hat{A}(355, 532)$, can be easily retrieved.

Mathematically, the depolarization ratio for pure pollen can be calculated using Eqs. (4), (5), and (7), as other variables are known or can be assumed. Nevertheless, we developed a retrieval method for this inverse model, so that it can be more easily applied to the real lidar measurements, especially for investigating the depolarization ratio with different values of the unknown $\hat{A}_{\text{pollen}}$. An iterate approach is used. In the first step, the depolarization ratio for pure pollen was assumed to be several different values (within the range

between 0.03 and 1), denoted as δ_x , in the simulator. The related pollen backscatter contribution ($\chi_{\text{pollen}}(532)$) inside the pollen layer can be retrieved using Eqs. (4) and (7). As its value depends on the assumed pollen depolarization ratio (δ_x), it can be expressed as $\chi_{\text{pollen}}(\delta_x, 532)$.

The relationship of $\hat{A}(355, 532)$ and $\chi_{\text{pollen}}(\delta_x, 532)$ was investigated using the parameter η (Eqs. 5 and 6). Examples of scatter plots using mean values of η and $\chi_{\text{pollen}}(\delta_x, 532)$ in the pollen layer for cases under the assumptions of $\delta_x = 0.1, 0.2, 0.3, 0.4$ and 0.5 are shown in Fig. 3. For these relationships, perfect linear fits (linear regression relationship) can be found and plotted as dotted lines in Fig. 3, following the simplified equation from Eqs. (5) and (6):

$$\eta(\chi_{\text{pollen}}(\delta_x, 532)) = a_1 \cdot \chi_{\text{pollen}}(\delta_x, 532) + a_0. \quad (8)$$

The fitting coefficient (a_1, a_0) values to determine the estimated parameter η are defined as in Eq. (5). Until this step of the inverse model, no assumption on the $\hat{A}_{\text{pollen}}$ was made; thus a_1 varies for different assumed values of δ_x . But a_0 is constant as the $\hat{A}_{\text{background}}$ is known. Theoretically, for each linear fit equation, $\chi_{\text{pollen}}(\delta_x, 532)$ values can range from 0 to 1, with 0 meaning no pollen and 1 meaning 100 % pollen in the observed aerosol particle population. Therefore, for each assumed δ_x , the η value for $\chi_{\text{pollen}}(\delta_x, 532) = 1$ can be defined as the value for pure pollen and denoted as $\eta_{\text{pure}}(\delta_x, 532)$.

In Sect. 3.1, the initial value of the backscatter-related Ångström exponent of pure pollen (denoted as $\hat{A}_{\text{pollen}}$) between 355 and 532 nm is 0, which results in an initial value of 1 for the parameter η . In this simulation, we assumed that the same value ($\hat{A}_{\text{pollen}} = 0$) should be retrieved; the goal was thus to find the value of 1 for η_{pure} . From previous results shown in Fig. 3, we can see that a δ_x value between 0.3 and 0.4 may result in $\eta_{\text{pure}} = 1$ (the black triangle in Fig. 3).

Hence, in the second step, more δ_x values in that range (0.3–0.4) were used in the simulation, and one can retrieve the relative value of $\eta_{\text{pure}}(\delta_x, 532)$ for each case. These values are presented in Fig. 4. The relationship between δ_x and $\eta_{\text{pure}}(\delta_x, 532)$ is not perfectly linear, but for these data inside the considered range, a good linear fit can be found with high

correlation coefficients ~ -1 . As there is noise in real lidar-measured profiles, two or more values of δ_x may be found as good solutions. However, after we introduce this additional second linear fit, only one solution will be retrieved in the end.

Finally, under the assumption of $\hat{A}_{\text{pollen}} = 0$, a pollen depolarization ratio of 0.35 was found, resulting in $\eta_{\text{pure}} = 1$ (shown by the black triangle in Fig. 4). This result is exactly the same as the initial value of the direct model, which validates the algorithm and provides the feasibility of using this inverse model to retrieve the pure pollen depolarization ratio values. A detailed flow chart of this inverse model is given in Fig. 5. Note that the values of δ_x can be chosen freely for values bigger than the background depolarization ratio and smaller than 1. This method can also be applied to the other two aerosol types (e.g. dust and non-dust aerosols), under the condition that the depolarization ratio of one aerosol type is the only unknown parameter, and other parameters are known or can be assumed, as long as both the depolarization ratio and the backscatter-related Ångström exponent of the two aerosol types are different.

3.3 Uncertainty study

The uncertainty study of this method is investigated in this section. The input parameters (i.e. initial values) of the direct model are defined in Sect. 3.1, with an 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 $\hat{A}_{\text{pollen}}$) were selected as different initial values for different uncertainty studies, which are clarified in each paragraph. The output of each direct model simulation was then used as the input for the inverse model.

Under ideal conditions, which means there is no noise on the input profiles for the inverse model, the depolarization ratio of pollen (depolarizing one) can be retrieved perfectly as long as the value is higher than the depolarization ratio of background aerosol (non-depolarizing one). δ_{pollen} of 0.04 has been tested, and the correct value was successfully retrieved. Note that for this case, the assumed values of δ_x should be selected as lower values (e.g. from 0.03). The more values of δ_x that are used in the inverse model, the better precision will be for the results, but a longer computation time is also needed. It is also possible to combine the first and second steps of the inverse model, by using many assumed values of δ_x (e.g. 0.032, 0.033, 0.034, ..., 0.98, 0.99) for the first step, at the cost of a long computation time.

In the cases presented, we assumed that the backscatter-related Ångström exponent of pure pollen between 355 and 532 nm to be used in the inverse model (denoted as $\hat{A}_{\text{pollen}}$) is 0, which was the same as the initial value ($\hat{A}_{\text{pollen}}$) of the direct model. But in reality, such information is not always available. Under different initial values of $\hat{A}_{\text{pollen}}$, there will

Figure 3. Scatter plots of mean values of η and $\chi_{\text{pollen}}(\delta_x, 532)$ in the pollen layer in five cases with assumed δ_x values. η is a parameter using the backscatter-related Ångström exponent between 355 and 532 nm (Eq. 6), and $\chi_{\text{pollen}}(\delta_x, 532)$ is the pollen backscatter contribution at 532 nm inside the pollen layer under a certain assumed pollen depolarization ratio value (δ_x is 0.1, 0.2, 0.3, 0.4, or 0.5). Linear regression lines are drawn by black dotted lines, with the fitting equation shown (Eqs. 5 or 8). The black triangle shows the ideal value: when χ_{pollen} is 1, η should be 1 ($\hat{A} = 0$).

Figure 4. Estimated parameter η_{pure} against the related assumed pollen depolarization ratio δ_x at 532 nm. η_{pure} is the $\eta(\chi_{\text{pollen}})$ value for the pure pollen (100 % pollen in the observed aerosol particle population, $\chi_{\text{pollen}} = 1$), where η is a parameter using the backscatter-related Ångström exponent between 355 and 532 nm (Eq. 6). The linear regression line is drawn by the black dotted line, with the fitting equation shown. The correlation coefficient (R^2) value is also given. The final result of 0.35 for pure pollen is found, resulting in $\eta_{\text{pure}} = 1$ (i.e. $\hat{A}_{\text{pollen}} = 0$) (by the black triangle).

Figure 5. Flow chart of the inverse model for the retrieval of the depolarization ratio value for pure pollen. The orange boxes are for the measured parameters (or simulated output from the direct model), the blue boxes for the assumptions/manual input and the green boxes for the estimations/calculations. A detailed description is in Sect. 3.2. The wavelength pair (λ_1, λ_2) is selected to be (355,532), (532,355), or (1064,532) in this study.

be a bias on the estimated values of pollen depolarization ratio if the assumed value is different (i.e. $\hat{A}_{\text{pollen}} \neq A_{\text{pollen}}$). For example, if the initial value $\hat{A}_{\text{pollen}}$ is 0.25 (i.e. $\eta_{\text{pure}} = 1.11$), but we keep the assumption of $\hat{A}_{\text{pollen}} = 0$ in the inverse model, the estimated pollen depolarization ratio is found to be 0.39 with a bias of 0.04 (shown in Fig. S3). The uncertainty due to the difference between the initial value of $\hat{A}_{\text{pollen}}$ and assumed $\hat{A}_{\text{pollen}}$ was simulated (shown in Fig. S4), where $\hat{A}_{\text{pollen}}$ is always assumed to be 0 in the inverse model. For initial values of $\hat{A}_{\text{pollen}} = \pm 0.5$ (i.e. bias of 0.5 on the assumed value of 0), relative uncertainties were assessed to be $\sim 30\%$. This uncertainty due to the difference of initial values of $\hat{A}_{\text{pollen}}$ and $\hat{A}_{\text{background}}$ was also investigated. The larger the difference between the two values ($\hat{A}_{\text{background}} - \hat{A}_{\text{pollen}}$), the smaller the uncertainty. For instance, if we use 3 (instead of 2) as the initial value of $\hat{A}_{\text{background}}$, the estimated pollen depolarization ratio is 0.37 (instead of 0.39), with a smaller bias for the above example.

Further on, we investigate the random uncertainty due to the noise on input lidar profiles, using the simulator and based on a Monte Carlo approach. The parameters for the six cases simulated earlier (as defined in Sect. 3.1, with values given in Table 1) are used again in this simulation, but

noises are additionally added, considering normal statistical distributions, which are introduced by a normal random generator (Fig. S1). The PDR and $\hat{A}$ are calculated from particle backscatter coefficients, so we only need to apply different noise levels to the particle backscatter coefficients in the direct model, and related PDR and $\hat{A}$ with noise can be retrieved. To simplify the problem, the initial noise levels for both backscatter coefficients at 355 and 532 nm were considered under the same assumptions. We defined “one group” as one draw of six simulated backscatter profiles with a certain noise level; these six backscatter profiles have a pollen OD of 0.002, 0.01, 0.02, 0.05, 0.1, and 1. For each statistical simulation, we used 200 draws (i.e. 200 groups of profiles). This uncertainty study was investigated in two parts:

- i. *Fix the input pollen depolarization ratio, and change the noise levels.* We used 0.35 as the initial pollen depolarization ratio. In the case of taking 10 % as the noise level on the backscatter coefficients, one group of six simulated profiles with noise is shown in Fig. 6. A pollen depolarization ratio of 0.354 was found for this group using the inverse model, with a bias of 0.004 compared to the initial value of 0.35. Similarly, pollen depolarization ratio values were retrieved for each of the 200 generated groups. These 200 values had a mean value

of 0.351 ± 0.009 ; thus an uncertainty of 0.009 (relative uncertainty of 2.6 %) was found. We changed the noise levels (e.g. 1 %, 10 %, 20 %, 40 %, and 60 %) on the backscatter coefficients by the normal random generator, and 200 draws were performed for each statistical simulation under each noise level. The uncertainties of the retrieved pollen depolarization ratio against the noise levels were assessed and are shown in Fig. 7a.

- ii. *Fix the noise level, and change the input pollen depolarization ratio.* In the second simulation, we keep 10 % as the noise level on the backscatter coefficients, and change the input pollen depolarization ratio values to 0.1, 0.2, 0.3, 0.4, and 0.5. Under each assumption, 200 draws were performed to derive the uncertainty values, which are reported in Fig. 7b. Relative uncertainties on the retrieved pollen depolarization ratio of 1.6 % to 2.8 % were found.

From the simulation results, small uncertainty and a good accuracy were found using this algorithm. Nevertheless, even with the introduced noise levels, these simulations were still performed under quasi-ideal conditions. For each simulated group, six cases were used to provide a wide range of values of χ_{pollen} (from ~ 0.05 to ~ 0.95), which leads to good constraints to find a fitting line for the regression relationship of χ_{pollen} and η (Eq. 6) (e.g. Fig. 3). If only three cases (with a pollen OD of 0.01, 0.02, and 0.05) were used for each group, uncertainties 2 to 5 times bigger were found. It is hard to give qualitative values for such an uncertainty study, but the wider range of χ_{pollen} values that are in the data set, the better the retrievals will be. The vertical resolution used here was 30 m (as the raw resolution of our lidar); and increasing the vertical resolution of the lidar would result in a smaller uncertainty in the simulation.

4 Results

4.1 Pollen grain and intense pollination period

During the 4-month campaign, 20 pollen types were observed and identified from the samples collected with the Burkard sampler, six from broadleaved trees, observed from the end of April to mid-June; three from coniferous trees, with a pollination period from mid-May to mid-June; and 11 from grass/weeds, observed mainly in July and August. Among them, birch (*Betula*), pine (*Pinus*), spruce (*Picea*), and nettle (*Urtica*) pollen were most abundant, contributing to more than 90 % of the total pollen load, regarding number concentrations. The surrounding forest is mixed in terms of the tree species, but the pollination periods of different dominant pollen types are distinct, as can be seen from the Burkard-observed number concentration of specific pollen types shown in Fig. 8a.

Microphotographs of pollen grains for the dominant pollen types are shown in Fig. 8b (photos taken from <https://www.paldata.org>, last access: 7 April 2020). Pine and spruce pollen belong to the *Pinaceae* family, which pollinate profusely and greatly contribute to the pollen counts. However, they are rarely considered as being allergenic. Their pollen grains are large due to their sacs or bladders, which make them easy to identify. Among winged grains, the body is sub-spheroidal to broadly ellipsoidal. The longest axis (sacci included) of *Pinus sylvestris* (Scots pine) pollen grains is 65–80 μm , while in *Picea abies* (Norway spruce) the axis is longer, 90–110 μm (Nilsson et al., 1977). Birch pollen can cause severe pollinosis and is recognized as one of the most important allergenic sources (D'Amato et al., 2007). Birch pollen grains are sub-oblate to oblate. *B. pubescens* pollen grains are 18–24 $\times$ 22–28 μm in size (Nilsson et al., 1977), and *B. pendula* (Silver birch) pollen grains are more or less the same size (personal communication with Sanna Pätsi from Aerobiology, University of Turku, 2020). Nettle is considered moderately allergenic, both in terms of skin tests and amount of exposure to the pollen in the air. Nettle (*Urtica dioica*) pollen grains are oblate spheroidal to spheroidal and are quite small, with a size of 13–17 $\times$ 15–20 μm (Nilsson et al., 1977). Information on the dominant pollen types is reported in Table 3, where the pollen season is defined using the 95 % method (Goldberg et al., 1988). The start of the season was defined as the date when 2.5 % of the seasonal cumulative pollen count was trapped and the end of the season when the cumulative pollen count reached 97.5 %.

Four intense pollination periods (IPPs) are defined considering the pollen seasons and the daily mean pollen concentration values of these four dominant pollen types (Table 3). A minimum value of 300 no. m^{-3} (for daily mean pollen concentration) was used as the threshold for the determination of IPP-1 and IPP-3, whereas a smaller threshold of 20 no. m^{-3} was used for IPP-2 and IPP-4. In addition, the availability of lidar measurements was considered for the IPP definition. IPP-1 and -2 are selected within the birch pollen season. During IPP-1, almost only birch pollen is observed (97 % contribution to number concentration), while during IPP-2, spruce pollen is additionally present in the air, with 14 % contribution. IPP-3 consists of two periods within the pine pollen season, separated by a few days with frequent low-level clouds (below 1 km) or rain, causing the relatively low pine pollen concentration between these two periods. IPP-4 is defined for the nettle pollen study for three separate short pollination periods in July and August.

4.2 Optical properties of pollen layer

4.2.1 Pollen layer

A pollen layer in the lidar measurements is defined as the lowest observed layer. The layer boundaries are determined using the gradient method (Bösenberg and Matthias, 2003;

Figure 6. Example of one group of six simulated profiles of (a) particle backscatter coefficient at 532 nm, (b) particle linear depolarization ratio at 532 nm, and (c) backscatter-related Ångström exponent between 355 and 532 nm. Profiles without noise are shown by dashed redlines, and ones with noise are shown by black lines. Noise levels on backscatter at both 355 and 532 nm were set to 10 %. Simulated results under six input pollen optical depth (OD) values of 0.002, 0.01, 0.02, 0.05, 0.1, and 1 (same as Fig. 1).

Table 3. (a) Dominant pollen types with their pollen season period, Latin name (taxa), and typical size. (b) Selected intense pollination periods (IPPs) and the presented dominant pollen types during each IPP. See more descriptions in Sect. 4.1.

(a) Dominant pollen types			
Pollen type	Pollen season in 2016 (dd.mm–dd.mm)	Taxa	The longest axis size (µm)*
Birch	29.04–26.05	<i>Betula</i>	22–28
Spruce	13.05–14.06	<i>Picea</i>	90–110
Pine	23.05–13.06	<i>Pinus</i>	65–80
Nettle	27.06–14.08	<i>Urtica</i>	15–20
(b) Selected intense pollination periods (IPPs)			
IPP	Period time in 2016 (dd.mm–dd.mm)	Pollen types (percentage of number concentration)	
IPP-1	05.05–09.05	Birch (97 %), other pollen (3 %)	
IPP-2	12.05–16.05	Birch (82 %), spruce (14 %), other pollen (4 %)	
IPP-3	23.05–25.05 & 28.05–03.06	Pine (95 %), other pollen (5 %)	
IPP-4	01.07–03.07 & 14.07–18.07 & 24.07–04.08	Nettle (75 %), other pollen (25 %)	

* Values from Nilsson et al. (1977).

Flamant et al., 1997; Mattis et al., 2008) based on the lidar-derived backscatter coefficient profile at 532 nm wavelength. A more detailed description of the layer definition method is described in Bohlmann et al. (2019). In this study, 2 h time-averaged lidar profiles are used to match the pollen sampler time resolution. The retrieved pollen layers are shown in Fig. 9a. With an overlap correction applied in this study, the lower limit for reliable backscatter profiles was about 600 m a.g.l. Statistical values of the pollen layer top height above ground level for the four IPPs were 1.5 ± 0.3 , 1.3 ± 0.3 , 1.3 ± 0.4 , and 1.2 ± 0.3 km, respectively (Fig. 9b). The lowest layer top height was found for the nettle pollen, belonging to herbaceous species. For the relatively larger spruce and

pine pollen, the layer top heights were lower compared to the smaller birch pollen.

4.2.2 Lidar-derived optical properties

Mean values of lidar-derived optical properties inside the detected pollen layers were retrieved (Table 4); these optical values represent the atmosphere with the presence of pollen (thus the mixture of pollen with other aerosols).

The lidar ratio (LR) at 532 nm and the LR at 355 nm for pollen layers were retrieved using the standard Raman method (Ansmann et al., 1990) during night-time measurements. The mean values are reported in Table 4, and box plots of LR at 532 nm and ratio of LRs are shown in Fig. 10a and b. Although the number of available profiles is limited,

Table 4. Lidar-derived optical values of pollen layer for the intense pollination periods (IPPs) (mean values $\pm$ standard derivation are given). LR: lidar ratio, PDR: particle linear depolarization ratio, $\text{\AA}$ bsc: backscatter-related Ångström exponent.

	Raman cases	LR 355 nm (sr)	LR 532 nm (sr)	All cases	PDR 532 nm	$\text{\AA}$ bsc 355–532 nm
IPP-1	10	54 ± 12	61 ± 8	37	0.08 ± 0.03	1.57 ± 0.43
IPP-2	7	71 ± 10	69 ± 4	15	0.25 ± 0.06	1.32 ± 0.61
IPP-3	13	66 ± 12	63 ± 14	46	0.14 ± 0.09	1.38 ± 0.57
IPP-4	15	63 ± 14	68 ± 11	45	0.04 ± 0.01	1.83 ± 0.43

Figure 7. Examples of estimated uncertainties (left y axis) and relative uncertainties (right y axis) on retrieved pollen depolarization ratio (DR_{pollen}) at 532 nm against (a) the applied noise levels on backscatter coefficient (Bsc), and (b) the initial input values of DR_{pollen} , using Monte Carlo method. The initial input value of DR_{pollen} is 0.35 for the example in (a). The noise level on backscatter coefficient (Bsc) is 10 % for the example in (b).

our results indicate that pollen consists of medium- to high-absorbing particles with values from 55 to 70 sr for all pollen types. For birch-dominant IPP-1 and nettle-dominant IPP-4, the LR of pollen layers at 532 nm is slightly larger than the LR at 355 nm. This behaviour is reversed for IPP-3 (pine-dominant) and IPP-2 (mixture of birch and spruce). However, no significant wavelength dependence can be determined on LR values accounting for the uncertainties.

The depolarization ratio was clearly enhanced when there were pollen grains in the air, and even higher depolarization ratios were observed with the presence of the more non-spherical spruce and pine pollen. Lidar-derived PDR values of detected pollen layers for the whole periods of each IPP

Figure 8. (a) Pollen concentration (2 h average) measured by the Burkard sampler at roof level. The main pollen types are shown according to the colour code. Defined intense pollination periods (IPPs) are shown by lines on the top. (b) Microphotographs of pollen grain: *Urtica* (nettle pollen), *Betula pendula* (birch pollen), *Pinus* (pine pollen), *Picea abies* (spruce pollen). Source: PalDat – a palynological database (<https://www.palдат.org>, last access: 7 April 2020).

are shown in Table 4 and Fig. 10c. This indicates that the depolarization ratio is the most proper indicator for pollen type. The extinction-related (not shown in this study) and backscatter-related Ångström exponent were also retrieved for pollen layers. The difference in the Ångström exponent for IPPs is much less evident, as the box plot of backscatter-related Ångström exponent between 355 and 532 nm shows (Fig. 10d). The use of the Ångström exponent to characterize pollen is quite delicate, as its value depends a lot on the background aerosol. Nevertheless, a clear tendency to a smaller Ångström exponent with an increasing depolarization ratio can be found, as is reported in Bohlmann et al. (2019). Thus under the same or similar background conditions, the Ångström exponent can be an indicator for pollen type. Even though we assumed that pollen grains were evenly distributed inside the pollen layer, a bigger pollen contribution in the aerosol mixture near the ground was observed.

Figure 9. (a) Pollen layer definition for four intense pollination periods (IPPs). (b) Box plot of pollen layer top heights during each IPP. The number of available profiles is given. Colours are related to the IPPs.

Figure 10. Box plots of (a) lidar ratio (LR) at 532 nm and (b) ratio of LR at 355 nm and LR at 532 nm during night-time measurements. Box plots of (c) particle linear depolarization ratio (PDR) and (d) backscatter-related Ångström exponent between 355 and 532 nm during all-day measurements. Mean values of the detected pollen layer for four IPPs are used. The horizontal line represents the median, the boxes the 25 % and 75 % percentiles, the whiskers the standard deviation, and the plus signs the outliers.

4.3 Estimation of optical properties for pure pollen from lidar observations

So far, we have retrieved the optical properties of the pollen layers, but the values for pure pollen are still unknown. In this section, the novel methodology presented in Sect. 3 is applied to the real lidar observations to estimate the optical properties for pure pollen particles.

4.3.1 Pollen optical properties at 532 nm

The method given in the inverse model module was applied to the real lidar observations in this section to retrieve the depolarization ratio at 532 nm for pure pollen. We assume that there are only pollen and non-depolarizing background aerosols in the air, which is reasonable because of the clean aerosol conditions at the measurement site.

For the first step, the depolarization ratio of pure pollen (δ_x) at 532 nm was assumed to be 0.2, 0.3, 0.4, or 0.5, and the depolarization ratio of non-pollen particles ($\delta_{\text{background}}$) at 532 nm was assumed to be 0.03. Under each assumption, we calculated the pollen backscatter coefficient during all IPPs and thus extract the related pollen backscatter contribution inside the pollen layer ($\chi_{\text{pollen}}(\delta_x, 532)$). Mean values of backscatter-related Ångström exponents between 355 and 532 nm inside the pollen layer were retrieved and denoted as $\hat{A}(355, 532)$. The relationship of $\hat{A}(355, 532)$ and $\chi_{\text{pollen}}(\delta_x, 532)$ of the pollen layers in each IPP was investigated using the parameter η (Eq. 6). The scatter plots using mean η and $\chi_{\text{pollen}}(\delta_x, 532)$ under different values of assumed δ_x (0.2, 0.3, 0.4, or 0.5) for IPP-1 and IPP-3 are given in the Supplement (Fig. S5 for IPP-1 and in Fig. S6 for IPP-3).

Based on results from the first step, in the second step, more δ_x values between 0.2 and 0.3 for IPP-1 (between 0.3 and 0.4 for IPP-3) were used for the calculations. Linear fitting lines were generated for the η and $\chi_{\text{pollen}}(\delta_x, 532)$ (Eq. 8) under each assumed δ_x . For these fitting lines, the η value for $\chi_{\text{pollen}}(\delta_x, 532) = 1$ was retrieved, denoted as $\eta_{\text{pure}}(\delta_x, 532)$ and reported in Fig. 11. η_{pure} presents the η values when the pollen contribution in the observed aerosol particle population is 100 %. Using these estimated $\eta_{\text{pure}}(\delta_x, 532)$ and δ_x , linear fits (shown by dotted lines in Fig. 11) can be assessed with high correlations.

Further on, the δ_x value which results in a certain value of $\eta_{\text{pure}}(\delta_x, 532)$ could be assumed as the depolarization ratio value of pure pollen. Under the assumption that the backscatter-related Ångström exponent of pure pollen (denoted as $\hat{A}_{\text{pollen}}$) between 355 and 532 nm is 0 (i.e. $\eta_{\text{pure}} = 1$), depolarization ratios 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 a pollen depolarization ratio of 0.24 and (b) for IPP-3 with a pollen depolarization ratio of 0.36. Good linear regression relationships are found for both cases, and two things should be highlighted: (1) $\hat{A}_{\text{pollen}}$ is 0 (i.e. $\eta_{\text{pure}} = 1$) for 100 % pollen in the observed aerosol particle popula-

Figure 11. Estimated η_{pure} against the related assumed pollen depolarization ratio δ_x at 532 nm for IPP-1 (in green) and IPP-3 (in blue). Linear regression lines are drawn by dotted lines, with fitting equations shown. The correlation coefficient (R^2) values are also given. η is a parameter using backscatter-related Ångström exponents between 355 and 532 nm (Eq. 6), and η_{pure} is the estimated η value for $\chi_{\text{pollen}}(\delta_x) = 1$ (i.e. the pollen contribution in the observed aerosol particle population is 100 %) (Eq. 8). The final results for pure pollen are shown by the black triangles. η_{pure} values of 0.82 and 1.22 (i.e. backscatter-related Ångström exponent of -0.5 and 0.5) are shown by horizontal dotted red lines.

tion (i.e. $\chi_{\text{pollen}} = 1$); and (2) without pollen in the air (i.e. $\chi_{\text{pollen}} = 0$), the backscatter-related Ångström exponent of non-pollen particles ($\hat{A}_{\text{background}}$) between 355 and 532 nm can be calculated, resulting in 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). There are no values of the Ångström exponent for pure pollen in the literature, but this assumption ($\hat{A}_{\text{pollen}} = 0$) is almost realistic, as pollen grains are quite big and thus can be assumed to be wavelength-independent of the backscatter at wavelengths of 355 and 532 nm. For big particles such as dust, Mamouri and Ansmann (2014) reported extinction-related Ångström exponents between 440 and 675 nm, with values of -0.2 for coarse dust and 0.25 for total dust.

An uncertainty study was investigated based on the method described 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 to be 0.24 for birch and 0.36 for pine for the uncertainty simulation; initial backscatter-related Ångström exponents of non-pollen particles between 355 and 532 nm were selected to be 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 $\hat{A}_{\text{pollen}}$ in the direct model and assumed $\hat{A}_{\text{pollen}}$ in the inverse mode were both selected to be 0. Estimated uncertainties were found to be 2.4 % for birch

and 2.9 % for pine (Table 5). Note that the different initial input values of $\hat{A}_{\text{pollen}}$ may introduce important additional bias. If we assume the true value of $\hat{A}_{\text{pollen}}$ is between -0.5 and 0.5 (i.e. values of η_{pure} from 0.82 to 1.22, shown by dotted red 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. The optical properties of pure pollen are lacking in the literature. Cao et al. (2010) measured the linear depolarization ratio of different pollen types in an aerosol chamber, by disseminating 2 g of the selected pollen; they determined a linear depolarization ratio at 532 nm for paper birch of 0.33 and for Virginia pine of 0.41. These values are higher than what we retrieved in this study, but it has to be kept in mind that these two experiments were conducted in quite different environments and conditions.

The retrieval of depolarization ratios for pure spruce or pure nettle pollen was not possible with this data set. During IPP-2, there was always a mixture of birch and spruce pollen with a variable mixing rate; in addition, the number of available measurements is limited. For nettle pollen, we have observed relatively small depolarization ratio values, together with a small variation, which makes the separation more challenging.

4.3.2 Pollen optical properties at 1064 and 355 nm

A similar study was performed to investigate the relationship between backscatter-related Ångström exponents between 532 and 1064 nm ($\hat{A}(1064, 532)$) and the pollen backscatter contribution at 532 nm; here we use another parameter η' (Eq. 6), which is a function of $\hat{A}(1064, 532)$, for the total particle backscatter. From the earlier simulations, we found out that the pollen backscatter contribution at 532 nm ($\chi_{\text{pollen}}(532)$) is proportional to the parameter η' , considering Eq. (5) using the wavelength pair of $\lambda_1 = 1064$ and $\lambda_2 = 532$.

The inverse model was applied for several assumed pollen depolarization ratios at 532 nm (ranging from 0.2 to 0.6), and no values of $\eta' = 1$ (i.e. $\hat{A}(1064, 532)_{\text{pollen}} = 0$) were found (Figs. S5, S6, S7). This result may be due to the fact that the laser beam at longer wavelengths would be more sensitive to bigger particles (pollen). Thus, there is some wavelength dependence on the backscattering between 532 and 1064 nm. The backscatter-related Ångström exponent of non-pollen particles between 532 and 1064 nm, denoted as $\hat{A}_{\text{background}}(532, 1064)$, can be calculated using Eq. (5) and the fitting equations in Figs. S5b and S6b, considering no pollen in the air (i.e. $\chi_{\text{pollen}} = 0$). A $\hat{A}_{\text{background}}(532, 1064)$ value of 1.0 or 1.1 (i.e. $\eta' = 0.50$ or 0.46) was estimated for IPP-1 or IPP-3, respectively. Considering the previously estimated depolarization ratio at 532 nm for pure birch (pine) pollen of 0.24 (0.36), the related η' was found to be 0.58 (0.69), corresponding to the value of ~ 0.8 (~ 0.5) for the backscatter-related Ångström exponent between 532 and 1064 nm. The extinction-related Ångström exponent is characterized mainly by the particle size, whereas

Table 5. Linear depolarization ratios for pure pollen. The assumption that the backscatter-related Ångström exponent between 355 and 532 nm for pollen ($\hat{A}_{\text{pollen}}$) should be 0 was applied for this study. The uncertainty on $\hat{A}_{\text{pollen}}$ was not taken into account for the standard deviation shown here, which may introduce non-negligible additional bias. For example, under the assumption of $\hat{A}_{\text{pollen}} \pm 0.5$, the range of depolarization ratio values is 0.19 to 0.27 for birch pollen and 0.26 to 0.44 for pine pollen. See more details in Sect. 4.3.

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

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 IPP-1 (a) and IPP-3 (b). η is a parameter using the 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 (Eqs. 5 or 8). The correlation coefficient (R^2) is also given. The size denotes the total pollen concentrations measured by the Burkard sampler at 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 the Supplement (Figs. S5 and S6).

the backscatter-related Ångström exponent depends on both the particle size and the refractive index (e.g. Amiridis et al., 2009; Giannakaki et al., 2010). Veselovskii et al. (2015) reported that the backscatter-related Ångström exponent between 355 and 532 nm is more sensitive to the refractive index, compare to the one between 532 and 1064 nm. In a study of Asian dust, Hofer et al. (2020) showed a larger range of values (−0.5 to 1.8) for the 355–532 nm backscatter-related Ångström exponent compared to the 532–1064 nm one (0.1 to 1.4).

A depolarization ratio at 355 nm can also be estimated, as pollen backscatter at both 355 and 532 nm should be the same under the assumption that the backscatter-related Ångström exponent between 355 and 532 nm for pure pollen is 0. The pollen backscatter contribution at 355 nm ($\chi_{\text{pollen}}(355)$) was calculated using the lidar-derived particle backscatter coefficient at 355 nm. The inverse model was applied here for the backscatter-related Ångström exponent between 355 and 532 nm ($\hat{A}(532, 355)$) and pollen backscatter contribution at

355 nm, using a third parameter $\frac{1}{\eta}$ (as in Eq. 6, a function of $\hat{A}(532, 355)$), which is proportional to the pollen backscatter contribution at 355 nm, considering Eq. (5) using the wavelength pair of $\lambda_1 = 532$ and $\lambda_2 = 355$. Here $\hat{A}$ is the backscatter-related Ångström exponent between 355 and 532 nm, for the total particle backscatter coefficient. Under different values of assumed pollen depolarization ratio at 355 nm ($\delta_{x,355}$) from 0.1 to 0.4, linear correlations were found for $\frac{1}{\eta}$ and $\chi_{\text{pollen}}(\delta_{x,355}, 355)$ (Fig. S8). Values for $\frac{1}{\eta_{\text{pure}}}(\delta_{x,355})$ for 100 % pollen backscatter contribution at 355 nm are reported in Fig. 13, against related $\delta_{x,355}$. Finally, the pollen depolarization ratios of 0.17 and 0.30 at 355 nm were found for IPP-1 (birch) and IPP-3 (pine), respectively (Table 5). Cao et al. (2010) found smaller values, with a linear depolarization ratio at 355 nm of 0.08 for paper birch and of 0.20 for Virginia pine.

The particle linear depolarization ratio at 355 nm can be calculated by using the pollen depolarization ratio at 355 nm. Mean values of depolarization ratio of pollen layers for IPP-1

Figure 13. Estimated $\frac{1}{\eta}_{\text{pure}}(\delta_{x,355})$ against the related assumed pollen depolarization ratio at 355 nm ($\delta_{x,355}$) for IPP-1 (in green) and IPP-3 (in blue). Linear regression lines are drawn by dotted lines, with fitting equations shown. The correlation coefficient (R^2) values are also given. $\frac{1}{\eta}$ is a parameter using the backscatter-related Ångström exponent between 355 and 532 nm (Eq. 6), and $\frac{1}{\eta}_{\text{pure}}$ is the estimated $\frac{1}{\eta}$ value for $\chi_{\text{pollen}}(\delta_{x,355}, 355) = 1$. The final results for pure pollen are shown by the black triangles. Results are under the assumption that the backscatter-related Ångström exponent between 355 and 532 nm for pure pollen is 0.

Figure 14. Box plots of estimated particle linear depolarization ratio (PDR) at 355 nm. Mean values of the detected pollen layer for all IPPs are used. The horizontal line represent the median, the boxes the 25 % and 75 % percentiles, the whiskers the standard deviation, and the plus signs the outliers. Results are under the assumption that the backscatter-related Ångström exponent between 355 and 532 nm for pure pollen is 0.

and IPP-3 were retrieved and are shown in Fig. 14. For both periods, PDR values at 355 nm are relatively smaller than the ones at 532 nm.

Uncertainty values for pollen depolarization ratios and particle linear depolarization ratio at 355 nm are not given in this paper, as these estimations were under the assumption that the backscatter-related Ångström exponent between 355 and 532 nm for pure pollen is 0 and are based on previously retrieved pollen depolarization ratios at 532 nm. More uncertainty sources should be considered for the uncertainty study, and it is complicated to give qualitative values. Nevertheless, a wavelength dependence seems to be found for depo-

larization values when pollen is present, which may be a key parameter for pollen recognition and characterization. Thus, depolarization ratios at different wavelengths are needed to identify different pollen types.

5 Summary and conclusions

We have defined lidar-derived properties for pure pollen based on a 4-month pollen campaign, which was performed during May to August 2016 at the Kuopio station in Eastern Finland. This station is part of the European Aerosol Research Lidar Network (EARLINET). Twenty types of pollen were observed and identified by a Burkard sampler, among which birch (*Betula*), pine (*Pinus*), spruce (*Picea*), and nettle (*Urtica*) pollen were the most abundant, contributing more than 90 % of the total pollen load, regarding number concentrations. Four intense pollination periods (IPPs) were defined considering the pollen seasons and the daily mean pollen concentration values.

Mean values of lidar-derived optical properties in the pollen layer were used to characterize pollen for each IPP. We found that lidar ratio (LR) values range from 55 to 70 sr for all pollen types, indicating that pollen consists of medium- to high-absorbing particles. No significant wavelength dependence could be determined on LR values using LRs at 355 and 532 nm, regarding the uncertainties. The wide range of LRs suggests that the LR alone is not a suitable parameter to discriminate between different pollen types. Nonetheless, we showed that the depolarization ratio is the most proper indicator for pollen and, further, the pollen type, as the depolarization ratio was enhanced when there was pollen in the air, and an even higher depolarization ratio was observed with the presence of the more non-spherical spruce and pine pollen. The Ångström exponent could be used to classify different pollen types only under the same or similar background conditions, as its value depends a lot on the background aerosols.

As the main results, we provide a novel method for the characterization of pure pollen particles. We present an algorithm to estimate the depolarization values for pure pollen, under the assumption that the backscatter-related Ångström exponent between 355 and 532 nm should be zero for pure pollen, as pollen grains are quite large and can be assumed to be wavelength-independent at these two wavelengths. This algorithm was first tested and validated through a simulator of synthetic lidar profiles (including a direct model and an inverse model module). 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 of pure pollen particles at 532 nm was assessed, resulting in 0.24 ± 0.01 and 0.36 ± 0.01 for birch and pine pollen, respectively. The un-

certainty on the assumed backscatter-related Ångström exponent of pure pollen will introduce non-negligible bias in addition, as discussed in Sect. 4.3.1. Pollen optical properties at 1064 and 355 nm were also estimated based on the retrieved pollen depolarization ratio at 532 nm. Pollen depolarization ratios of 0.17 and 0.30 at 355 nm were found for birch and pine pollen, respectively. The depolarization values show a wavelength dependence for pollen. This can be the key parameter for pollen detection and characterization. Also, a wavelength dependence on the backscatter between 532 and 1064 nm was found, with the value of the backscatter-related Ångström exponent of ~ 0.8 (~ 0.5) for pure birch (pine) pollen between 532 and 1064 nm. Based on simulations in this study, we found that depolarization ratios at 355 nm and 1064 nm provide valuable information for pollen study; thus more multi-wavelength lidar studies on the depolarization characterization of atmospheric pollen are necessary. The presented novel algorithm and the estimated optical properties for pure pollen in this study provide a good method for pollen characterization and classification. Currently, the CALIPSO (Cloud-Aerosol Lidar with Orthogonal Polarization) aerosol type classification scheme includes seven tropospheric aerosol types (Kim et al., 2018; https://www-calipso.larc.nasa.gov/resources/calipso_users_guide/data_summaries/vfm/index_v420.php, last access: 7 April 2020), in which pollen (or biogenic aerosols in general) is excluded. Such ground-based lidar measurements also provide the possibility of implementing a new aerosol type in the CALIPSO classification scheme, for example using the depolarization ratio at 532 nm. This method can also be applied to other aerosol mixtures (e.g. dust and non-dust aerosols) to retrieve the particle linear depolarization ratio related to aerosol types, under the condition that the depolarization ratio of one aerosol type is the only unknown parameter, and other parameters are known or can be reasonably well approximated. Note that the two constraints mentioned in Sect. 3.1 should be considered: both the depolarization ratio and the backscatter-related Ångström exponent of the two aerosol types should be different.

Data availability. Lidar data are available upon request from the authors, and data quick-looks are available on the PollyNET website (<http://polly.tropos.de/>, last access: 25 June 2020).

Supplement. The supplement related to this article is available online at: <https://doi.org/10.5194/acp-20-15323-2020-supplement>.

Author contributions. XS analysed the data, developed the algorithm and the simulator, and wrote the paper. XS, EG, MK, and SR conceptualized and finalized the methodology. XS and SB performed the lidar data analysis. AS analysed the pollen samples. MK and EG initiated and managed the project. MF, AR, AL, and

MK participated in the measurement campaign. All authors were involved in the editing of the paper, interpretation of the results, and discussion of the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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Aerosol particle depolarization ratio at 1565 nm measured with a Halo Doppler lidar

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Abstract. The depolarization ratio is a valuable parameter for lidar-based aerosol categorization. Usually, the aerosol particle depolarization ratio is determined at relatively short wavelengths of 355 nm and/or 532 nm, but some multi-wavelength studies including longer wavelengths indicate strong spectral dependency. Here, we investigate the capabilities of Halo Photonics StreamLine Doppler lidars to retrieve the particle linear depolarization ratio at the 1565 nm wavelength. We utilize collocated measurements with another lidar system, PollyXT at Limassol, Cyprus, and at Kuopio, Finland, to compare the depolarization ratio observed by the two systems. For mineral-dust-dominated cases we find typically a slightly lower depolarization ratio at 1565 nm than at 355 and 532 nm. However, for dust mixed with other aerosol we find a higher depolarization ratio at 1565 nm. For polluted marine aerosol we find a marginally lower depolarization ratio at 1565 nm compared to 355 and 532 nm. For mixed spruce and birch pollen we find a slightly higher depolarization ratio at 1565 nm compared to 532 nm. Overall, we conclude that Halo Doppler lidars can provide a particle linear depolarization ratio at the 1565 nm wavelength at least in the lowest 2–3 km above ground.

1 Introduction

Aerosols and their interactions with clouds remain the largest source of uncertainty in the Earth's radiative budget (IPCC, 2013). Remote sensing measurements with lidars enable continuous long-term observations of the vertical distribution of aerosol particles and clouds in the atmosphere, providing valuable information for improving our understanding of the global climate system (e.g. Illingworth et al., 2015). Information on the vertical distribution of aerosols is highly important also for the aviation industry in case of hazardous aerosol emissions from for example volcanic eruptions (Hirtl et al., 2020).

Lidar measurements of aerosol optical properties at multiple wavelengths can be used to categorize elevated aerosol layers into different types such as mineral dust, smoke, marine aerosol or volcanic ash (e.g. Baars et al., 2017; Papaioannopoulos et al., 2018). One of the most important parameters for such aerosol typing is the depolarization ratio, which enables distinguishing spherical and non-spherical particles from each other (e.g. Burton et al., 2012; Baars et al., 2017). Furthermore, the depolarization ratio can be used to quantify the contributions of different aerosol types to elevated layers (Mamouri and Ansmann, 2017). It is essential also for esti-

mating vertical profiles of cloud condensation nuclei (CCN) and ice-nucleating particle (INP) concentrations from remote sensing observations (Mamouri and Ansmann, 2016).

Currently, the particle linear depolarization ratio is most commonly measured at relatively short wavelengths of 355 and/or 532 nm (e.g. Illingworth et al., 2015; Baars et al., 2016), though some lidar systems are capable of depolarization ratio measurement at longer wavelengths of 710 and 1064 nm (e.g. Freudenthaler et al., 2009; Burton et al., 2012). For instance, Burton et al. (2012) used the ratio of depolarization ratios at 1064 and 532 nm as part of their aerosol typing procedure. However, to our knowledge, the aerosol particle depolarization ratio has not been reported at wavelengths longer than 1064 nm.

Previous studies on the spectral dependency of the depolarization ratio between 355 and 1064 nm have shown a steep decrease in the depolarization ratio from 532 to 1064 nm for elevated biomass burning aerosols (Burton et al., 2012, 2015; Haarig et al., 2018; Hu et al., 2019). On the contrary, mineral dust layers present an increasing depolarization ratio with increasing wavelength (Groß et al., 2011; Burton et al., 2015) or a relatively weak maximum at 532 nm (Freudenthaler et al., 2009; Burton et al., 2015; Haarig et al., 2017). For some aerosol types, such as marine aerosol (Groß et al., 2011) and volcanic ash (Groß et al., 2012), no spectral dependency was observed. However, volcanic ash mixed with boundary layer aerosol was observed with a clearly lower depolarization ratio at 355 nm than at 532 nm (Groß et al., 2012).

The spectral dependency of the depolarization ratio has been attributed largely to the shape of the size distribution of polarizing aerosol particles. In smoke layers, the depolarization signal is probably due to non-spherical soot aggregates, which are in the size range of 100 nm to hundreds of nanometres and thus do not produce a large depolarization ratio at 1064 nm (Burton et al., 2015; Haarig et al., 2018; Hu et al., 2019). Recently, Gialitaki et al. (2020) modelled smoke as near-spherical submicron particles and found good agreement with the observed spectral dependency of the depolarization ratio. On the other hand, mineral dust contains significant amounts of coarse-mode particles ($>1\ \mu\text{m}$ in diameter), which can explain the large depolarization ratio also observed at the 1064 nm wavelength (Freudenthaler et al., 2009; Groß et al., 2011; Burton et al., 2015; Haarig et al., 2017). In aged dust layers, the faster removal of supermicron particles is thought to result in the depolarization ratio peaking at 532 nm (Freudenthaler et al., 2009; Groß et al., 2011; Burton et al., 2015; Haarig et al., 2017). In other words, spectral analysis of the depolarization ratio could permit more in-depth diagnosis of coarse-mode polarizing aerosol.

Halo StreamLine Doppler lidars are commercially available fibre-optic systems that operate at a wavelength of 1565 nm and can be equipped with a cross-polar receiver channel for measuring the depolarization ratio (Pearson et al., 2009). Over the last few years these lidars have become widely used in wind and turbulence studies (e.g. Päsche et

Figure 1. Locations of Vehmasmäki and Limassol measurement sites.

al., 2015; Vakkari et al., 2015; Tuononen et al., 2017; Manninen et al., 2018). Additionally, depolarization ratio measurements by Halo lidars have been used to study cloud and precipitation phase (e.g. Achtert et al., 2015).

Now, recently developed post-processing (Vakkari et al., 2019) allows the utilization of significantly weaker signals from Halo Doppler lidars than previously. Therefore, the main aim of this paper is to assess the capabilities of Halo Doppler lidars in providing particle linear depolarization ratio measurements at the 1565 nm wavelength. To do so, we utilize collocated Halo Doppler lidar and multiwavelength Raman lidar PollyXT observations during two measurement campaigns, where different polarizing aerosols were observed. Overall, the comparison indicates that Halo Doppler lidars can add another wavelength at 1565 nm to studies on the spectral dependency of the particle linear depolarization ratio, at least in the lowest 2–3 km above ground.

2 Materials and methods

Here we use data from two measurement campaigns where a Halo Photonics Doppler lidar and a PollyXT Raman lidar were collocated: at Kuopio, Finland, from 9 to 16 May 2016 and at Limassol, Cyprus, from 21 April to 22 May 2017. The campaigns represent quite different environments (Fig. 1) and enable the comparison of the depolarization ratio at 1565 nm by the Halo instrument to the depolarization ratio at 355 and 532 nm from PollyXT for a range of aerosol types. Furthermore, the campaigns were equipped with different devices of the Halo and PollyXT designs, and thus potential differences between instrument individuals can be investigated.

The Vehmasmäki site (62.738° N, 27.543° E; 190 m a.s.l.) in Kuopio is a rural location surrounded by boreal forest (Bohlmann et al., 2019). The focus of the campaign in

May 2016 was to investigate the capability to characterize the optical properties of airborne pollen with the multiwavelength Raman lidar PollyXT (Bohlmann et al., 2019). Here, we utilize 1 week of collocated measurements to compare Halo depolarization at 1565 nm to PollyXT during a spruce and birch pollination episode.

Limassol (34.675° N, 33.043° E; 22 m a.s.l.) is located at the southern shore of Cyprus in the eastern Mediterranean. Measurements at Limassol were part of the Cyprus Clouds Aerosol and Rain Experiment (CyCARE; Ansmann et al., 2019) and were performed as a collaboration between the Cyprus University of Technology (CUT), Limassol, and the Leibniz Institute for Tropospheric Research (TROPOS), Leipzig. During April–May, several Saharan dust episodes were observed at Limassol in addition to the regional aerosol.

2.1 Halo Doppler lidar

Halo Photonics StreamLine lidars are commercially available 1565 nm pulsed Doppler lidars equipped with a heterodyne detector (Pearson et al., 2009). Halo StreamLine lidars emit linearly polarized light, and the optical path is constructed with fibre-optic components, which can be equipped with a cross-polar receiver channel. The cross-polar channel is implemented through a fibre-optic switch between the normal receiver path and path with a fibre-optic polarizer. Thus, the measurement of the co- and cross-polar signals is not simultaneous but consecutive in vertically pointing mode. For instance, if the integration time per ray is set to 7 s, then co-polar signal is collected for 7 s and then cross-polar signal is collected during the next 7 s.

For research purposes, the most commonly used variants of StreamLine lidars are StreamLine, StreamLine Pro and StreamLine XR. The StreamLine and the more powerful StreamLine XR lidars enable full hemispheric scanning. The Streamline Pro is designed without moving parts on the outside, which limits the scanning to a cone of 20° from vertical. All StreamLine variants can be used for depolarization ratio measurements, but an important difference between XR and other StreamLine versions is that the XR background noise level cannot be determined as accurately in the near range as for the non-XR versions (Vakkari et al., 2019). This difference is attributed to the more sensitive amplifier used in the StreamLine XR (Vakkari et al., 2019).

In this study we utilize vertically pointing measurements only from two StreamLine Pro systems. The operating specifications of these systems are given in Table 1. StreamLine lidars send and receive pulses through a single lens, which avoids issues with overlap and leads to a minimum range of 90 m due to impact of the outgoing pulse. At Vehmasmäki, we focused on boundary layer aerosol and set integration time per ray to 7 s and telescope focus to 2000 m. At Limassol, we expected to encounter elevated aerosol layers frequently and set the integration time per ray to 11.5 s and telescope focus to infinity. The integration time is set to bal-

Table 1. Specifications of Halo Doppler lidars used in this study.

Wavelength	1565 nm
Pulse repetition rate	15 kHz
Pulse energy	20 µJ
Pulse duration	0.2 µs
Nyquist velocity	20 m s ⁻¹
Sampling frequency	50 MHz
Velocity resolution	0.038 m s ⁻¹
Points per range gate	10
Range resolution	30 m
Maximum range	9600 m
Lens diameter	8 cm
Lens divergence	33 µrad
Telescope	monostatic optic-fibre coupled

ance between signal strength and good enough time resolution for retrievals of turbulent properties.

Halo StreamLine lidars provide three parameters along the beam direction: radial Doppler velocity, signal-to-noise ratio (SNR) and attenuated backscatter (β), which is calculated from SNR taking into account the telescope focus. For a coherent Doppler lidar attenuated backscatter is obtained as

$$\beta(z) = A \frac{\text{SNR}(z)}{T_f(z)}, \quad (1)$$

where z is the range from the instrument, A incorporates system-specific constants and $T_f(z)$ is the telescope focus function, which includes range correction (Frehlich and Kavaya, 1991; Pentikäinen et al., 2020).

A background check to determine range-resolved background noise level is performed automatically once per hour. The raw signal from atmospheric measurement is then divided by this noise level in the firmware and returned as SNR (see Vakkari et al., 2019). We post-processed SNR according to Vakkari et al. (2019), which ensures that both co- and cross-polar SNR have an unbiased noise level; i.e. SNR is 0 when there is no signal (cf. Manninen et al., 2016). Furthermore, the post-processing is essential to be able to further reduce the instrumental noise floor by averaging the SNR (Vakkari et al., 2019). After post-processing SNR, β is calculated with Eq. (1).

Halo depolarization ratio

We estimate the instrumental uncertainty in Halo StreamLine SNR from the standard deviation of SNR in the cloud- and aerosol-free part of the profile. Given the long wavelength and low pulse energy, no contribution from molecular scattering is observed in the signal. At 1565 nm the molecular backscatter coefficient is about $1.9 \times 10^{-8} \text{ m}^{-1} \text{ sr}^{-1}$ at mean sea level, using mean values for the atmospheric number density taken from the US Standard Atmosphere, 1976 (COESA 1976). The two-way atmospheric transmittance at 1565 nm is still 0.9994 at 2 km altitude above a lidar situated at mean sea

Figure 2. Profiles at Limassol, Cyprus, on 2 May 2017 at 12:08 UTC. **(a)** Co- and cross-polar SNR. A liquid cloud at approx. 800 m a.g.l. results in full attenuation of signal. Below cloud layer aerosol signal is visible. Above 1 km variability in SNR is due to instrumental noise only. **(b)** The same as panel (a) but limited to lowest 1 km a.g.l. **(c)** Ratio of cross-polar SNR to co-polar SNR up to 1 km a.g.l. calculated from profiles in panel (a). Error bars represent uncertainty due to instrumental noise estimated from SNR at > 1 km a.g.l. in panel (a).

level. Hence, the measured depolarization ratio can be safely assumed to represent the particle linear depolarization ratio.

In Fig. 2a, consecutive co- and cross-polar SNR profiles are presented, where aerosol signal is observed up to 800 m above ground level (a.g.l.) and a liquid cloud base is observed at 840 m a.g.l. In liquid cloud the signal attenuates quickly, and above 1 km the profiles represent instrumental noise only. We use the measurements above 1 km to calculate standard deviations of co-polar SNR (σ_{co}) and cross-polar SNR (σ_{cross}). In Fig. 2c, the raw depolarization ratio (δ^*) is calculated simply as the ratio of cross-polar SNR to co-polar SNR, and uncertainty is estimated from σ_{co} and σ_{cross} by Gaussian error propagation.

The construction of Halo StreamLine lidars does not include a calibrator for depolarization channel, unlike PollyXT lidars for instance (Engelmann et al., 2016). Furthermore, the user cannot change the optical path to include a calibrator or check the depolarizing effects of the individual components. Therefore, we are limited to evaluating the Halo depolarization ratio at liquid cloud base.

Spherical cloud droplets do not polarize the directly backscattered radiation, and thus non-zero δ^* at liquid cloud base is an indication of incomplete extinction (or bleed-through) in the lidar internal polarizer. However, measurement of δ^* at cloud base can be biased by signal saturation or changes in cloud properties between co- and cross-polar measurement. Furthermore, multiple scattering results in increasing depolarization signal inside a liquid cloud (e.g. Liou and Schotland, 1971). This increase in in-cloud δ^* is clearly seen in Fig. 2c: at cloud base δ^* is 0.0102, and at the next gate 30 m deeper inside the cloud δ^* has increased to 0.0116.

The magnitude of the multiple scattering effect on depolarization ratio depends on both cloud and lidar properties

(e.g. Donovan et al., 2015). In Halo StreamLine lidars the instrument telescope design has a matched field of view and divergence of $33 \mu\text{rad}$ (Table 1), and consequently the effect is small: in Fig. 2c δ^* increases by 0.0014 in 30 m. For instance, for the system modelled by Donovan et al. (2015) in-cloud multiple scattering increases the depolarization ratio from 0 to 0.05 in approx. 50 m. Nevertheless, to minimize the effect of multiple scattering we only consider δ^* at the cloud base for the determination of the average bleed-through and use measurements in several clouds.

For low-level clouds, we have observed saturation of the co-polar signal in the more powerful StreamLine XR instruments. Signal saturation at liquid cloud base is readily identified as a non-linear relationship between co- and cross-polar SNR. For the measurement cases analysed here, we did not observe indications of saturation. Furthermore, we note that δ^* at cloud base should be determined with as high a time resolution as possible to ensure that both co- and cross-polar measurements represent the same part of the cloud. In practice, integration time is kept constant during a measurement campaign and was set as a compromise between temporal resolution and signal strength. We mitigate the effect of poor time resolution by choosing cases where cloud base remains at the same altitude (within lidar resolution) for some tens of minutes and thus one can assume temporal homogeneity. No vertical smoothing is applied in calculating δ^* , as the signal at cloud base is strong compared to the aerosol signal. Finally, it should be noted that, especially in higher latitudes, it is not always trivial to find purely liquid phase clouds. Typically, mixed-phase clouds can be distinguished by the depolarizing features of ice crystals. That is, in the histogram of δ^* at cloud base a secondary peak with higher δ^* than liquid clouds would occur, which was not the case for our study.

To characterize the Halo polarizer bleed-through, we determined the δ^* at liquid cloud base during both campaigns (Fig. 3). During the campaign at Limassol, we determined δ^* at cloud base on 25 April and on 2 May 2017. From the distribution in Fig. 3a, the bleed-through is 0.011 ± 0.007 (mean $\pm$ standard deviation). At Vehmasmäki, we utilized clouds on 13, 14 and 16 May 2016 as shown in Fig. 3b. At Vehmasmäki, the estimated bleed-through is 0.016 ± 0.009 (mean $\pm$ standard deviation). The mean cloud base δ^* values observed for these two systems in Fig. 3 are well in line with our experience with these and five other StreamLine and StreamLine XR systems in Finland, where cloud base δ^* typically ranges from 0.01 to 0.02.

We attribute the spread in the distributions in Fig. 3 mostly to variability of the clouds at the measurement sites and to the fact that co- and cross-polar measurements are consecutive and not simultaneous. Given that the cross-polar measurement channel is constructed with fibre-optic technology, we do not expect changes in the performance of the polarizer. This is also our experience with Halo systems in Finland since 2016, but we recommend to check the bleed-through monthly or after an instrument is moved to a new location.

Figure 3. Ratio of cross-polar SNR to co-polar SNR at liquid cloud base measured with Halo Doppler lidar. **(a)** Distribution of cloud base δ^* at Limassol. **(b)** Distribution of cloud base δ^* at Vehmasmäki.

Considering the large natural variability of the depolarization ratio (e.g. Illingworth et al., 2015; Baars et al., 2016) we find the spread of observations in Fig. 3 tolerable. The standard deviation in Fig. 3 is included in the uncertainty calculation of the Halo depolarization ratio.

We account for the observed bleed-through (B) in calculating Halo particle linear depolarization ratio (δ_{1565}) as

$$\delta = \frac{\text{SNR}_{\text{cross}} - B \cdot \text{SNR}_{\text{co}}}{\text{SNR}_{\text{co}}}, \quad (2)$$

where SNR_{co} and $\text{SNR}_{\text{cross}}$ are the observed co- and cross-polar SNR, respectively. Uncertainty in $\text{SNR}_{\text{cross}}$ corrected for bleed-through (i.e. numerator in Eq. 2) is estimated as

$$\sigma_{\text{cross}, B} = \sqrt{\sigma_{\text{cross}}^2 + (B \cdot \text{SNR}_{\text{co}})^2 \cdot \left(\frac{\sigma_B^2}{B^2} + \frac{\sigma_{\text{co}}^2}{\text{SNR}_{\text{co}}^2} \right)}, \quad (3)$$

where σ_B is standard deviation of the distribution in Fig. 3. Finally, uncertainty in δ_{1565} taking into account instrumental noise and uncertainty in bleed-through is estimated as

$$\sigma_{\delta} = |\delta| \sqrt{\frac{\sigma_{\text{cross}, B}^2}{(\text{SNR}_{\text{cross}} - B \cdot \text{SNR}_{\text{co}})^2} + \frac{\sigma_{\text{co}}^2}{\text{SNR}_{\text{co}}^2}}. \quad (4)$$

2.2 PollyXT

PollyXT is a multiwavelength Raman lidar capable of depolarization ratio measurement at one or two wavelengths depending on instrument configuration (Baars et al., 2016; Engelmann et al., 2016). PollyXT emits simultaneously 355, 532 and 1064 nm wavelength pulses at a repetition frequency of 20 Hz. All PollyXT lidars are built with the same design, but there are small differences in the number of receiver channels equipped in each individual system. A detailed description of PollyXT design is given by Baars et al. (2016) and Engelmann et al. (2016).

At Vehmasmäki, PollyXT was configured with elastic backscatter channels (355, 532 and 1064 nm); Raman-shifted channels at 387, 407 and 607 nm; and a cross-polar channel

at 532 nm (Bohlmann et al., 2019). Due to the biaxial construction of emission and detection units, complete overlap is reached at 800–900 m a.g.l. (Engelmann et al., 2016), and thus only measurements >800 m a.g.l. are utilized for this study (Bohlmann et al., 2019). The original spatial resolution is 30 m and temporal resolution 30 s for the Vehmasmäki system (Bohlmann et al., 2019).

At Limassol, PollyXT operated the same receiver channels as the Vehmasmäki system had and additionally a cross-polar channel at 355 nm, together with a near-range telescope with 355 and 532 nm receiver channels. The near-range channels enable retrieval of optical properties down to 150 m a.g.l. (Engelmann et al., 2016). Raw spatial resolution is 7.5 m and temporal resolution is 30 s.

During night-time, the Raman method (Ansmann et al., 1992) is used to retrieve aerosol optical properties from the raw signals. For daytime measurements, the method of Klett (1981) can be utilized. Here, we present only measurements when the Raman method was applied. The calibration of the depolarization ratio was performed at both Vehmasmäki and Limassol using the so-called $\Delta 90^\circ$ method (Freudenthaler, 2016), and the relative uncertainty in the particle linear depolarization ratio was estimated to be 10 %.

2.3 Auxiliary data

Air mass history was estimated with the Hybrid Single-Particle Lagrangian Integrated Trajectory model, HYSPLIT (Stein et al., 2015). HYSPLIT was run through the READY website (Rolph et al., 2017) using the NCEP Global Data Assimilation System (GDAS) meteorology at 0.5° horizontal resolution. Back trajectories of 96 h arriving at the elevation of aerosol layers of interest were calculated.

3 Results

In this section we analyse observations of dust, marine and pollen aerosols during the Limassol and Vehmasmäki campaigns, where said aerosol types were observed simultaneously with Halo and PollyXT lidars. Dust and marine aerosols were observed during the Limassol campaign in the eastern Mediterranean, and pollen was observed during the Vehmasmäki campaign in a boreal forest region in Finland. We conclude this section with an overall comparison of the depolarization ratio measurements with the two instruments.

3.1 Elevated dust layers

3.1.1 Limassol 21 April 2017

Right at the beginning of Halo measurements at Limassol on 21 April 2017, several elevated layers were observed as seen in Fig. 4. Although Halo can observe elevated layers up to 6 km a.g.l. on this day, the signal is too weak to retrieve their depolarization ratio. This is clearly visible in the uncer-

Figure 4. Limassol 21 April 2017 measurements with Halo Doppler lidar. (a) Time series of co-polar SNR at 300 s integration time. (b) Time series of cross-polar SNR at 300 s integration time. (c) Uncertainty in the depolarization ratio. (d) Particle linear depolarization ratio filtered with $\sigma_\delta < 0.05$.

tainty in the Halo depolarization ratio in Fig. 4c. At 300 s integration time (i.e. 10 min of alternating co- and cross-polar measurement), the depolarization ratio can be determined up to 1–1.5 km a.g.l. with $\sigma_\delta < 0.05$ on this day (Fig. 4d). The depolarization ratio can be retrieved also for the relatively strong elevated layer at 3 km a.g.l. during the morning hours (Fig. 4d).

Increasing both temporal and spatial averaging enables the utilization of some of the weaker signals. Figure 5 presents profiles of the Halo and PollyXT depolarization ratio, where both are averaged over 1.5 h (20:00–21:30 UTC) and smoothed vertically with a 300 m running mean. In the lowest layer < 1 km a.g.l., practically no difference is observed in the depolarization ratio at the different wavelengths. Back-trajectory calculations (Fig. 6) indicate this layer to be mostly regional air from eastern Mediterranean, and the relatively large lidar ratio is in the range of observations of smoke or smoke mixed with dust (e.g. Groß et al., 2011; Baars et al., 2016). On the other hand, for the layer from 1.5 km to 2 km a.g.l. a clear increase in δ with increasing wavelength is observed. For this layer air mass history indicates origins over northern Africa (Fig. 6), and the lidar ratio (42 ± 4 at 355 nm, 47 ± 5 at 532 nm) is in the range of dust (Ansmann et al., 2011). For this layer the mean ($\pm$ standard deviation) δ values at 355, 532 and 1565 nm are 0.19 ± 0.008 , 0.23 ± 0.008 and 0.29 ± 0.008 , respectively. Above 2 km a.g.l., the uncertainty in δ at 1565 nm increases rapidly and is not used for quantitative analysis here.

3.1.2 Limassol 27 April 2017

Stronger elevated aerosol layers were observed at Limassol on 27 April 2017. On this day, the depolarization ratio can be retrieved by Halo up to 3 km a.g.l. (Fig. 7). For an averaging period of 01:25–02:30 UTC, the depolarization ratio

Figure 5. Averaged profiles at Limassol on 21 April 2017 at 20:00–21:30 UTC. All profiles have been smoothed by a 300 m running mean. (a) Backscatter by PollyXT (wavelengths 355–1064 nm) and attenuated backscatter by Halo (1565 nm). (b) Particle linear depolarization ratio. Error bars represent measurement uncertainty. (c) Lidar ratio. For PollyXT β_{355} , β_{532} , LR_{355} and LR_{532} , a near-range telescope is used for data < 900 m a.g.l.

Figure 6. Back trajectories of 96 h arriving at Limassol on 21 April 2017 at 21:00 UTC.

is retrieved for the elevated layer at 1600–2200 m a.g.l. For this layer, the depolarization ratio at 1565 nm is 0.30 ± 0.005 , which is a little lower than for the shorter wavelengths: 0.36 ± 0.01 at 355 nm and 0.34 ± 0.002 at 532 nm, respectively. For this layer, the air mass history indicates southerly origins.

On the same day (27 April 2017) at 19:00–20:00 UTC, the depolarization ratio can be retrieved from the surface up to 2.6 km a.g.l. (Fig. 8). In the lowest 500 m, the depolarization ratio at 1565 nm is clearly higher than at the shorter wavelengths, suggesting a mixture of larger mineral dust particles with smaller particles of a lower depolarization ratio. For the layer at 1500–2500 m a.g.l., practically no wavelength dependency is observed for the depolarization ratio, indicating that backscatter at all wavelengths is dominated by the same aerosol. The layer-averaged depolarization ratios are 0.31 ± 0.006 , 0.33 ± 0.005 and 0.32 ± 0.008 at 355, 532 and 1565 nm, respectively. The high depolarization ratio and li-

Figure 7. Limassol 27 April 2017 measurements with Halo Doppler lidar. (a) Time series of co-polar SNR at 300 s integration time. (b) Time series of cross-polar SNR at 300 s integration time. (c) Uncertainty in the depolarization ratio. (d) Particle linear depolarization ratio filtered with $\sigma_\delta < 0.05$.

Figure 8. Averaged profiles at Limassol on 27 April 2017 at 19:00–20:00 UTC. All profiles have been smoothed by a 300 m running mean. (a) Backscatter by PollyXT (wavelengths 355–1064 nm) and attenuated backscatter by Halo (1565 nm). (b) Particle linear depolarization ratio. Error bars represent measurement uncertainty. (c) Lidar ratio. For PollyXT β_{355} and β_{532} , a near-range telescope is used for data < 900 m a.g.l.

dar ratio of 47 ± 5 at 355 nm (38 ± 3 at 532 nm) indicate almost pure dust (Ansmann et al., 2011; Baars et al., 2016). Air mass history, on the other hand, indicates northerly or north-westerly origins at both 2 km a.g.l. and at the surface (Fig. 9).

3.2 Polluted marine aerosol

On 20 May 2017 at Limassol, a very low aerosol depolarization ratio is observed throughout the day as seen in Fig. 10. During the morning and afternoon liquid clouds are observed, but during the evening Raman retrievals with PollyXT were possible. Figure 11 presents Halo depolar-

Figure 9. Back trajectories of 96 h arriving at Limassol on 27 April. Back trajectories arriving at 19:00 and 02:00 UTC are included.

Figure 10. Limassol 20 May 2017 measurements with Halo Doppler lidar. (a) Time series of co-polar SNR at 300 s integration time. (b) Time series of cross-polar SNR at 300 s integration time. (c) Uncertainty in the depolarization ratio. (d) Particle linear depolarization ratio filtered with $\sigma_\delta < 0.05$.

ization ratio profiles averaged for the duration of the PollyXT retrieval at 19:54–21:30 UTC. For the surface layer (up to 1 km a.g.l.), a small decrease in the depolarization ratio with increasing wavelength is observed. The layer-averaged depolarization ratios are 0.03 ± 0.01 , 0.015 ± 0.002 and 0.009 ± 0.003 at 355, 532 and 1565 nm, respectively. The layer-averaged lidar ratio at 355 nm is 39 ± 4 sr, whereas the lidar ratio at 532 nm is very noisy at 47 ± 35 sr. The low depolarization ratio is typical of marine aerosol, smoke and pollution (Groß et al., 2011; Illingworth et al., 2015). The 355 nm lidar ratio lies between the values reported for marine aerosol and smoke (Illingworth et al., 2015).

Above 1 km a.g.l., an optically thin aerosol layer is observed (Fig. 11). Halo indicates a higher depolarization ratio for this layer than at the surface, but the signal is so weak that the uncertainty in the depolarization ratio at 1565 nm becomes very large (Fig. 11b). Back trajectories arriving

Figure 11. Averaged profiles at Limassol on 20 May 2017 at 19:55–21:30 UTC. All profiles have been smoothed by a 300 m running mean. (a) Backscatter by PollyXT (wavelengths 355–1064 nm) and attenuated backscatter by Halo (1565 nm). (b) Particle linear depolarization ratio. Error bars represent measurement uncertainty. (c) Lidar ratio. For PollyXT β_{355} , β_{532} , LR_{355} and LR_{532} , a near-range telescope is used for data <900 m a.g.l.

Figure 12. Back trajectories of 96 h arriving at Limassol on 20 May 2017 at 21:00 UTC.

over Limassol at 21:00 UTC indicate different but mostly northerly origins for the air mass at 500 m and at 2 km a.g.l. (Fig. 12).

3.3 Pollen in boreal forest

On 15 May 2016, substantial amounts of spruce and birch pollen were observed at Vehmasmäki with both an in situ sampler and the PollyXT lidar (Bohlmann et al., 2019). The presence of more polarizing spruce pollen (Bohlmann et al., 2019) in the boundary layer is observed also with Halo lidar as seen in Fig. 13d. However, the backscatter (Fig. 14a) is low compared to the case studies presented for Limassol, and the low signal results in significant noise in the lidar ratio (Fig. 14c).

Comparing the depolarization ratios measured with Halo and PollyXT (Fig. 14b) shows a nearly constant depolariza-

Figure 13. Vehmasmäki 15 May 2016 measurements with Halo Doppler lidar. (a) Time series of co-polar SNR at 350 s integration time. (b) Time series of cross-polar SNR at 350 s integration time. (c) Uncertainty in the depolarization ratio. (d) Particle linear depolarization ratio filtered with $\sigma_\delta < 0.05$.

Figure 14. Averaged profiles at Vehmasmäki on 15 May 2016 at 19:00–21:00 UTC. All profiles have been smoothed by a 300 m running mean. (a) Backscatter by PollyXT (wavelengths 355–1064 nm) and attenuated backscatter by Halo (1565 nm). (b) Particle linear depolarization ratio. Error bars represent measurement uncertainty. (c) Lidar ratio.

tion ratio at 1565 nm, while the depolarization ratio at 532 nm decreases with height. At 1565 nm, the Halo signal is probably dominated by pollen grains, which are tens of micrometres in diameter. At the 355 and 532 nm wavelengths, the backscatter is increasing with height (Fig. 14a), and thus the decreasing depolarization ratio at 532 nm may reflect an increasing fraction of signal from non-pollen aerosol with increasing height. For the layer from 800 m to 1 km a.g.l. in Fig. 14, the mean depolarization ratios are 0.236 ± 0.009 and 0.269 ± 0.005 at 532 and 1565 nm, respectively.

Figure 15. Comparison of particle linear depolarization ratio at different wavelengths. Observations represent 30 m vertical resolution and have been smoothed by a 300 m running mean. To reduce scatter, a stricter uncertainty threshold is applied, and only data for $\sigma_\delta < 0.01$ (at the 1565 nm wavelength) are included. The mean is calculated at intervals of 0.025 on the x axis, and error bars indicate standard deviation. (a) Depolarization ratio at 1565 nm (Halo) vs. depolarization ratio at 532 nm (PollyXT) at Limassol. (b) Depolarization ratio at 1565 nm vs. depolarization ratio at 355 nm (PollyXT) at Limassol. (c) Depolarization ratio at 532 nm vs. depolarization ratio at 355 nm at Limassol. (d) Depolarization ratio at 1565 nm vs. depolarization ratio at 532 nm at Vehmasmäki.

3.4 Overview of depolarization ratio wavelength dependency

An overall comparison of the depolarization ratio at different wavelengths for the Limassol and Vehmasmäki campaigns is presented in Fig. 15, where the Halo vertical resolution of 30 m has been smoothed with a 300 m running mean. The original time resolution observations by Halo have been averaged to match the temporal resolution of PollyXT Raman retrievals (ranging from 45 min to 2 h).

In Fig. 15a, three regions can be observed in the scatterplot. For $\delta_{532} < 0.05$, δ_{1565} matches very closely with the shorter wavelength. For δ_{532} ranging from 0.05 to 0.25, δ_{1565} is systematically larger than δ_{532} . For $\delta_{532} > 0.3$, δ_{1565} is lower than the depolarization ratio at the shorter wavelength. A very similar pattern is present in Fig. 15b: for $\delta_{355} < 0.05$, δ_{1565} matches δ_{355} closely; for δ_{355} ranging from 0.05 to 0.25, δ_{1565} is larger than δ_{355} ; and for $\delta_{355} > 0.3$, δ_{1565} is lower than δ_{355} . Similar regions appear even when comparing the two shorter wavelengths (Fig. 15c): for $\delta_{355} < 0.05$, δ_{532} is lower than δ_{355} ; for δ_{355} ranging from 0.1 to 0.3, the depolarization ratio is on average equal on both wavelengths; and for $\delta_{355} > 0.3$, δ_{532} is lower than δ_{355} .

Figure 15a–c also show similar correlations between the depolarization ratios at different wavelengths. Therefore, bearing in mind the similar patterns in all three scatterplots in Fig. 15a–c, we consider the scatter to originate mainly from the atmospheric aerosol properties rather than in instrumental

effects. For instance, any bias in the estimated bleed-through in the Halo polarizer would show up as bias in Fig. 15a and b. However, such bias is not present in the cases when δ_{355} and/or δ_{532} are low.

Considering the sources at Limassol during the campaign, the higher δ_{1565} for intermediate depolarization ratios ranging from 0.1 to 0.25 likely represents mixtures of dust with other aerosol types. A mixture of coarse, polarizing dust with less polarizing and smaller aerosol would result in the observed spectral dependency of the depolarization ratio. For aged-dust-dominated cases, lower depolarization ratios at longer wavelength could be due to the faster removal of coarse particles compared to submicron aerosol (e.g. Burton et al., 2015). In any case, the observed wavelength dependency in Fig. 15a–c for large δ suggests that, for dust-dominated cases, smaller particle sizes have, on average, a higher depolarization ratio at Limassol.

Another type of polarizing aerosol, i.e. pollen, was observed with a collocated Halo and PollyXT at Vehmasmäki (Bohmann et al., 2019). Comparatively low signal levels, together with 800 m minimum range for the PollyXT system at Vehmasmäki (Bohmann et al., 2019), reduce the amount of data available for comparison of Halo and PollyXT depolarization ratio during the campaign (Fig. 15d). During this campaign, the depolarization ratio at 1565 nm is a little larger than at 532 nm, but the difference is small compared to the scatter observed at Limassol.

A further look into the distribution and spectral dependency of the depolarization ratio at Limassol is presented in Fig. 16. In Fig. 16a and b, the 2D histograms of the depolarization ratio show that both the 532 and 1565 nm wavelengths present a bi-modal distribution below 1 km a.g.l. In other words, there are also less polarizing aerosols frequently present in the lowest 1 km in addition to dust and dusty mixtures with a depolarization ratio > 0.2 . However, above about 1.5–2 km a.g.l., almost all retrievals indicate dust or dusty mixtures. Note that the vertical extent of the data is limited by the sensitivity of the Halo instrument, as Fig. 16a and b are limited to cases when both wavelengths are available.

In Fig. 16c and d, the ratio of depolarization ratios at 1565 and 532 nm exhibits a clear height dependency. Above about 1.5 km a.g.l., the majority of the observations present a lower depolarization ratio at 1565 nm than at 532 nm, while below 1.5 km a.g.l. the depolarization ratio is higher at the longer wavelength. In previous studies (Freudenthaler et al., 2009; Groß et al., 2011; Burton et al., 2015; Haarig et al., 2017), a lower depolarization ratio at longer wavelengths has been attributed to faster removal of coarse-mode dust. However, our observations indicate the presence of a small coarse-mode, probably mineral, dust for sub-1.5 km aerosols most of the time at Limassol.

Figure 16. 2D histograms of the particle linear depolarization ratio and height at Limassol using 30 m vertical resolution smoothed by a 300 m running mean. Only data for $\sigma_\delta < 0.01$ (at the 1565 nm wavelength) are included. (a) Depolarization ratio at 532 nm. (b) Depolarization ratio at 1565 nm. (c) Ratio of depolarization ratios at 1565 and 532 nm. (d) Same as panel (c) but scaled with number of observations at each height.

4 Discussion

The majority of aerosol depolarization ratio measurements have been carried out at relatively short wavelengths (355 and 532 nm) with only a few previous studies investigating the spectral dependency including 710 nm (Freudenthaler et al., 2009; Groß et al., 2011) and/or 1064 nm (Freudenthaler et al., 2009; Burton et al., 2012, 2015; Haarig et al., 2017, 2018; Hu et al., 2019). In this study we have for the first time determined aerosol particle depolarization ratios at a wavelength of 1565 nm.

From an instrumental point of view, the Halo Doppler lidar depolarization ratio seems to be of comparable quality to the PollyXT depolarization ratio when the aerosol signal is strong. However, Halo has a much less powerful laser than PollyXT, which significantly limits the range of usable signal. On the other hand, Halo Doppler lidars are capable of independent operation for months and are therefore suitable for operational use in meteorological measurement networks.

The integration time and range gate length are adjustable in Halo firmware, and prolonging these parameters would increase the sensitivity of the system. However, high spatial and temporal resolution are preferable for utilizing the Doppler capabilities of Halo lidars. Inspecting the internal polarizer performance at liquid cloud base also requires a higher resolution. Overall, the configuration of a Halo Doppler lidar needs to be considered individually for the aims of each measurement campaign.

The spectral dependency that we observed for the 355, 532 and 1565 nm particle linear depolarization ratio agrees reasonably well with previous spectral analyses for similar aerosol types as seen in Table 2 and Fig. 17. For the mineral dust depolarization ratio, both decreasing and increasing

Figure 17. Particle linear depolarization ratio as a function of wavelength for dust observations in Table 2. Additionally, spectral dependency modelled with MOPSMAP based on the OPAC database for desert dust (Koepke et al., 2015; Gasteiger and Wiegner, 2018) and AERONET inversion by Toledano et al. (2019) are included.

trends with increasing wavelength have been observed previously (Table 2). This is the case for our observations at Limassol as well, though on average δ_{1565} tends to be a little lower than δ_{532} (Fig. 16). Probably, the spectral dependency of the mineral dust depolarization ratio depends on both the age of the dust and the origin of the dust. The spectral dependency of the depolarization ratio modelled with MOPSMAP (Gasteiger and Wiegner, 2018) for desert dust aerosol from the OPAC database (Koepke et al., 2015) agrees reasonably well with the Saharan dust case on 21 April 2017 in this study (Fig. 17). On the other hand, the sun-photometer-based retrieval by Toledano et al. (2019) for long-range transported Saharan dust over Barbados indicates a slightly lower depolarization ratio of 0.19 at 1640 nm compared to this study at 1565 nm (Fig. 17). The lower depolarization ratio at 1640 nm over Barbados is reasonable considering the much longer transport compared to this study.

Wavelength-dependent changes in the mineral dust depolarization ratio are small compared to elevated smoke layers, which can help to distinguish between these two aerosol types (Burton et al., 2012). For elevated smoke, a strong decrease in the depolarization ratio has been reported from >0.20 at short wavelengths to $\delta < 0.05$ at 1064 nm (Burton et al., 2015; Haarig et al., 2018; Hu et al., 2019). Thus, adding a depolarization ratio measurement at 1565 nm can provide added value to the commonly used measurements at the 355 and 532 nm wavelengths.

For marine aerosols, the depolarization ratio is small and has practically no spectral dependency (Groß et al., 2011), which is what we observed at Limassol. For the mixture of spruce and birch pollen at Vehmassäki, the differences in the depolarization ratio at 532 and 1565 nm are small.

5 Conclusions

In this paper we report for the first time remote sensing measurements of the atmospheric aerosol particle linear depolar-

Table 2. Spectral dependency of depolarization ratio for dust, marine aerosol and pollen.

	Time and origin	Depolarization ratio				
		355 nm	532 nm	710 nm	1064 nm	1565 nm
This study, Limassol	21 Apr 2017 20:00–21:30; Saharan dust	0.19 ± 0.008	0.23 ± 0.008			0.29 ± 0.008
	27 Apr 2017 01:25–02:33; dust (Egypt)	0.36 ± 0.01	0.34 ± 0.002			0.30 ± 0.005
	27 Apr 2017 at 19:00– 20:00; dust (Turkey)	0.31 ± 0.006	0.33 ± 0.005			0.32 ± 0.008
Haarig et al. (2017)	Barbados 2013, 2014; Saharan dust	0.252 ± 0.030	0.280 ± 0.020		0.225 ± 0.022	
Burton et al. (2015)	US 2014; Saharan dust	0.209 ± 0.015	0.304 ± 0.005		0.270 ± 0.005	
	Mexico Chihuahua 2013; local dust	0.225 ± 0.041	0.373 ± 0.014		0.383 ± 0.006	
Groß et al. (2011)	Cabo Verde 2008; Saharan dust	0.24–0.27	0.29–0.31	0.36–0.40		
Freudenthaler et al. (2009)	Morocco 2006; Saharan dust	0.24–0.28	0.31 ± 0.03	0.26–0.30	0.27 ± 0.04	
This study, Limassol	20 May 2017 at 19:55– 21:30; polluted marine	0.03 ± 0.01	0.015 ± 0.002			0.009 ± 0.003
Groß et al. (2011)	Cabo Verde 2008; marine	0.02 ± 0.01	0.02 ± 0.02			
This study, Vehmasmäki	15 May 2016 19:00–21:00; spruce and birch pollen		0.236 ± 0.009			0.269 ± 0.005

ization ratio at a wavelength of 1565 nm. Using observations at liquid cloud base we have been able to characterize the Halo Doppler lidar polarizer bleed-through with sufficient accuracy to obtain useful depolarization ratio measurements; uncertainty in the bleed-through is propagated to the depolarization ratio measurement. A comparison of two different Halo Doppler lidar systems with two PollyXT systems during collocated measurements at Limassol, Cyprus, and Kuopio – Vehmasmäki, Finland, shows good agreement between the lidar systems. The agreement between the instruments is remarkably good considering the large wavelength difference: the PollyXT depolarization ratio is retrieved at 355 and/or 532 nm. However, given the much lower laser energy in Halo Doppler lidars, it is not surprising that the vertical extent of the usable depolarization ratio is much lower than for PollyXT.

For relatively fresh mineral dust, we find particle linear depolarization ratios at 1565 nm ranging from 0.29 to 0.32, which is in good agreement with previous observations, including measurements at the 710 and 1064 nm wavelengths (Freudenthaler et al., 2009; Groß et al., 2011; Burton et al., 2015; Haarig et al., 2017). For polluted marine aerosol we observed a very low depolarization ratio of 0.009

at 1565 nm with a small decrease with increasing wavelength. The spruce and birch pollen depolarization ratio has been characterized only recently at 532 nm (Bohlmann et al., 2019). Our measurements indicate a slightly higher depolarization ratio of 0.27 at 1565 nm compared to 0.24 at 532 nm. Overall, our results indicate that Halo Doppler lidars can add another wavelength at 1565 nm to studies on the spectral dependency of the particle linear depolarization ratio, at least in the lowest 2–3 km above ground.

For aerosol typing, adding a particle linear depolarization ratio at 1565 nm to shorter wavelengths can help to distinguish biomass burning aerosols from dust, as a much stronger spectral dependency has been observed for elevated biomass burning aerosols than for dust (e.g. Haarig et al., 2017, 2018; Hu et al., 2019). In case there is prior knowledge of prevailing aerosols, such as transport of volcanic ash, even stand-alone particle linear depolarization ratio measurements with Halo Doppler lidars can probably provide useful information for aerosol typing.

Data availability. Processed lidar data are available upon request from the authors. Level 0 PollyXT observations are available at <https://polly.tropos.de/> (Polly NET, 2020, last access: 18 Au-

gust 2020). Trajectory model HYSPLIT and GDAS meteorological data are available at <https://www.ready.noaa.gov/HYSPLIT.php> (ARL, 2020, last access: 18 August 2020).

Author contributions. Conceptualization and formal analysis were carried out by VV, while investigation was carried out by HB, SB, JB, MK, RM and EJO'C. Data curation was carried out by VV, HB, SB, MK and EJO'C. VV wrote the original draft, which was reviewed and edited by VV, HB, SB, MK and EJO'C.

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Lidar depolarization ratio of atmospheric pollen at multiple wavelengths

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Abstract. Lidar observations during the pollen season 2019 at the European Aerosol Research Lidar Network (EARLINET) station in Kuopio, Finland, were analyzed in order to optically characterize atmospheric pollen. Pollen concentration and type information were obtained by a Hirst-type volumetric air sampler. Previous studies showed the detectability of non-spherical pollen using depolarization ratio measurements. We present lidar depolarization ratio measurements at three wavelengths of atmospheric pollen in ambient conditions. In addition to the depolarization ratio detected with the multiwavelength Raman polarization lidar Polly^{XT} at 355 and 532 nm, depolarization measurements of a co-located Halo Doppler lidar at 1565 nm were utilized. During a 4 d period of high birch (*Betula*) and spruce (*Picea abies*) pollen concentrations, unusually high depolarization ratios were observed within the boundary layer. Detected layers were investigated regarding the share of spruce pollen to the total pollen number concentration. Daily mean linear particle depolarization ratios of the pollen layers on the day with the highest spruce pollen share are 0.10 ± 0.02 , 0.38 ± 0.23 and 0.29 ± 0.10 at 355, 532 and 1565 nm, respectively, whereas on days with lower spruce pollen share, depolarization ratios are lower with less wavelength dependence. This spectral dependence of the depolarization ratios could be indicative of big, non-spherical spruce pollen. The depolarization ratio of pollen particles was investigated by applying a newly devel-

oped method and assuming a backscatter-related Ångström exponent of zero. Depolarization ratios of 0.44 and 0.16 at 532 and 355 nm for the birch and spruce pollen mixture were determined.

1 Introduction

Pollen is an essential part of plant reproduction as it stores and transports genetic information of the plant. Because of the immobility of plants, pollen grains need to be transferred by other means, for example, wind or animals. Most grasses (*Poaceae*), conifers and around 10 %–20 % of flowering plants are wind pollinated (Ackerman, 2000). Typical sizes of anemophilous pollen grains range from 20 to 60 µm (Shukla et al., 1998), but also large pollen grains like pine (*Pinus*) or spruce (*Picea*) pollen with diameters around 100 µm are transported by wind. Because anemophilous plants produce pollen in very large numbers, they are almost omnipresent in the environment. Thus, it is not surprising that pollen has various effects on climate and human health. Millions of people suffer from pollen-triggered allergies and the number is expected to rise due to the changing climate (Schmidt, 2016). Increasing temperatures and higher CO₂ concentrations favor plant growth and pollen production. In regions with sufficient water resources, the grow-

ing seasons are therefore expected to become longer and the spatial distribution of allergenic species can be expected to change (Ziska and Beggs, 2012).

Pollen can be lifted up to several kilometers by turbulent mixing within the boundary layer. Airborne measurements such as during the Cooperative Convective Precipitation Experiment (CCOPE) near Miles City, Montana (USA), revealed the presence of pollen up to 4 km above ground with a considerable amount of pollen still observed at 2 km (Mandrioli et al., 1984). Once reached the top of the boundary layer, pollen can be transported by wind over thousands of kilometers (Rousseau et al., 2008; Skjth et al., 2007; Szczepanek et al., 2017). Dispersed in the atmosphere, it can affect the climate. By acting as ice-nucleating particles, pollen promotes the formation of clouds (von Blohn et al., 2005; Diehl et al., 2001, 2002). Furthermore, pollen grains can rupture and lead to a high number of smaller particles. Those pollen fragments are efficient cloud condensation nuclei (Steiner et al., 2015; Wozniak et al., 2018).

Considering the relevance of pollen for climate and environment and the increasing importance as health hazard, it is of high importance to study the pollen distribution and transport mechanism in the atmosphere. To further improve pollen transport and forecast models, validation measurements are needed. Previously, we have shown that in the absence of other non-spherical particles, light detection and ranging (lidar) measurements and especially the particle depolarization ratio can be used to track pollen grains in the atmosphere (Bohlmann et al., 2019). We also estimated the depolarization ratio at 532 nm for pure birch (*Betula*) and pine pollen under certain assumptions as 0.24 ± 0.01 and 0.36 ± 0.01 , respectively (Shang et al., 2020). However, due to the measurement setup in our earlier studies, the lowest layers of the atmosphere (below 800 m), which naturally contain most of the airborne pollen, could not be investigated. In this study, we extend our dataset by operating an upgraded version of our multiwavelength Raman lidar (Polly^{XT}) with four near-range channels at the same measurement site in Kuopio (Vehmasmki), Finland. The improved lidar setup allowed measurements down to around 350 m above ground level. Furthermore, the new system measures the depolarization ratio also at 355 nm, while depolarization measurements were limited to 532 nm in the previous campaign. To further improve the investigation of pollen depolarization properties, we assessed the depolarization measurements of a co-located Halo Doppler lidar, which provides information at 1565 nm (Vakkari et al., 2021). The measurement of the depolarization ratio at multiple wavelengths allows us to investigate its wavelength dependence. This could enable the distinction of pollen from other depolarizing aerosols, as the depolarization ratio at single wavelengths can be similar for different aerosol types.

Instrumentation and methods are presented in Sect. 2. The observation period is introduced in Sect. 3. Therein, we present a case study and comparison of different pollen con-

ditions during the period under study. Supporting measurement data, a statistical analysis, the discussion of the wavelength dependence and an estimation of pollen depolarization values are also given in same section.

2 Instrumentation and methods

Our measurement campaign was performed from 23 April to 28 August 2019 at the rural forest site in Vehmasmki (Kuopio), Finland (6244' N, 2733' E; 190 m above sea level). Vehmasmki is surrounded by broadleaved and coniferous forest. Dominant tree species include Silver birch (*Betula pendula*), Norway spruce (*Picea abies*) and Scots pine (*Pinus sylvestris*). The measurement site is permanently equipped with a Raman polarization lidar, a Halo Doppler lidar, multiple in situ instruments as well as standard meteorological instruments located on top of a measurement container. Furthermore, a 318 m tall mast provides measurements of temperature, relative humidity and wind at various levels from ground up to 300 m. During our measurement campaign, a Hirst-type spore trap was deployed on top of the container to measure the pollen concentration. In the following subsections, the instruments and pollen detection are introduced, and the layer detection method is explained.

2.1 Polly^{XT} and processing of lidar observations

The multiwavelength Raman polarization lidar Polly^{XT}-FMI (Engelmann et al., 2016) is a fully automated lidar instrument, which emits at three wavelengths (355, 532 and 1064 nm), and is able to detect at two rotational–vibrational Raman channels (387 and 607 nm) and one water vapor detection channel at 407 nm in addition to the elastic backscatter channels at the emission wavelengths. The cross-polarized signal can be detected at 355 and 532 nm, which allows the determination of the linear volume depolarization ratio (VDR) and linear particle depolarization ratio (PDR) at those two wavelengths. In addition to the far-field channels, Polly^{XT} includes a near-field unit with detection channels at 355, 387, 532 and 607 nm. The combination of near- and far-field channels allows the observation of continuous profiles down to about 120 m, the height of complete overlap of the near-field receiver. The initial vertical resolution of the acquired data is 7.5 m and the temporal resolution is 30 s. Further details on the instrument setup, principle and error propagation can be found in Engelmann et al. (2016). Near-real-time quicklooks are publicly accessible at the PollyNET website (<http://picasso.tropos.de/>, last access: 17 December 2020), which visualizes the measurements of all lidars belonging to PollyNET network (Baars et al., 2016).

The lidar data were analyzed using the Klett–Fernald method (Fernald, 1984; Klett, 1981) whenever the weather conditions were adequate (i.e., no low clouds or rain) and the signal quality was sufficient for deriving high-quality pro-

files. A constant lidar ratio of 60 sr was used to derive profiles of backscatter and extinction coefficients. The lidar ratio is based on measurements retrieved during nighttime using the Raman method (Ansmann et al., 1990) and the results for birch–spruce mixtures of our previous study (Bohlmann et al., 2019). Optical products retrieved from far- and near-field channels were merged to obtain the best possible profiles throughout the whole troposphere, enabling us to determine reliable profiles close to the ground. However, this is not the case for the 355 nm channel. Due to an overlap issue on the 355 nm far-range channel, reliable profiles at this wavelength were limited to 900 m above ground. The problem also affected the calculation of the depolarization ratio at 355 nm, which therefore could not be retrieved close to ground. To obtain high-quality depolarization measurements, calibration measurements are automatically performed three times a day using the $\Delta 90^\circ$ method (Freudenthaler, 2016; Freudenthaler et al., 2009). A calibration constant (V_λ^*) determined from two subsequent measurements with a difference of exactly 90° is used to calibrate the volume depolarization ratio. During the presented time period, quite stable calibration constants V^* of $1.85 \times 10^{-1} \pm 4.01 \times 10^{-3}$ at 355 nm and $6.90 \times 10^{-3} \pm 6.29 \times 10^{-5}$ at 532 nm were determined and applied to calibrate depolarization measurements. The profiles were temporally averaged in 2 h intervals to match with the time resolution of the pollen samples. Aerosol layers within the lowest 4 km of the atmosphere were detected using an automatic layer detection algorithm. The layer detection is based on the gradient of the backscattered signal at 1064 nm. The local maximum of the first derivative is considered to be the layer bottom, while the height of the local minimum of the first derivative corresponds to the layer top (Belegante et al., 2014). Less smoothed profiles with a vertical smoothing range of 127 m were used to determine the correct geometrical layer boundaries without losing layer depth. Higher smoothing with a vertical range of 367 m, in turn, was applied to calculate reasonable optical properties within the layer. Intensive optical properties (lidar ratios, linear particle depolarization ratios and Ångström exponents), commonly used for lidar-based aerosol characterization, were calculated for the automatically detected layers. Especially, the linear depolarization ratio and backscatter-related Ångström exponents (BÅE) are considered in this study. The linear depolarization ratio describes the change of the polarization state of the backscattered light. The strength of depolarization depends on the non-sphericity of the backscattering particles and the particle size relative to the wavelength (Sassen, 2005). The depolarization ratio increases with non-sphericity and can therefore be used to characterize the particle shape. The Ångström exponents describe the wavelength dependence of the scattering/extinction and relate to the particle size.

2.2 Halo Doppler lidar

In addition to the Polly^{XT} lidar, a Halo Photonics StreamLine Pro Doppler lidar (Pearson et al., 2009) is operated permanently at our measurement site. This lidar operates at a wavelength of 1565 nm with a pulse repetition rate of 15 kHz and a pulse energy of 20 μ J. The minimum range of the instrument is 90 m and the range resolution is 30 m; other operating specifications can be found in Vakkari et al. (2021). This Halo Doppler lidar is equipped with a cross-polar receiver channel, which enables consecutive measurement of co- and cross-polar signals in vertically pointing mode and thus the retrieval of the depolarization ratio at 1565 nm. In this study, an integration time of 7 s was used; i.e., the co-polar signal is collected for 7 s followed by the cross-polar signal collected during the next 7 s. In addition to vertically pointing measurement, a conical scan lasting 2.5 min was scheduled every 15 min to retrieve the horizontal wind profile.

The Halo Doppler lidar signal-to-noise ratio was post-processed according to Vakkari et al. (2019) and the depolarization ratio was processed according to Vakkari et al. (2021). The design of Halo Doppler lidars does not include a depolarization calibration unit nor does it allow us to add one. Therefore, depolarization measurements can only be calibrated using the depolarization ratio at liquid cloud base. As liquid cloud droplets do not polarize the backscattered light, non-zero depolarization ratios at cloud base indicate incomplete extinction, called bleed-through, in the internal polarizer (Vakkari et al., 2021). The bleed-through in the Halo internal polarizer was estimated from liquid cloud base observed at 1200–1400 m a.g.l. on 14 June at 00:30–03:30 UTC as 0.013 ± 0.006 . This estimate is very close to the estimate of 0.016 ± 0.009 from measurements with same instrument in 2016 (Vakkari et al., 2021). The depolarization ratio was corrected for the bleed-through according to Vakkari et al. (2021). The retrieved depolarization ratio can be considered as the linear particle depolarization ratio, since the contribution of molecular scattering at this long wavelength and low pulse energy is neglectable (Vakkari et al., 2021).

Vertical wind speed from the co-polar vertically pointing measurements and horizontal wind profile from the conical scans were used to calculate the turbulent kinetic energy (TKE) dissipation rate following O'Connor et al. (2010).

2.3 Pollen observations

The pollen information was obtained using a Hirst-type volumetric air sampler located on top of the measurement container, 4 m above ground. The sampler collects particles down to a size of 3.7 μ m (Muilenberg, 2003) onto an adhesive-coated tape which is manually analyzed afterwards. With this type of sampler, it is possible to determine pollen types and concentrations with 2 h time resolution. A more

Figure 1. Pollen micrographs from 18 May 2019 (a) and 19 May 2019 (b) at 09:00–11:00 UTC. Examples of birch and spruce pollen grains are marked with green and red arrows, respectively (source: Biodiversity Unit of the University of Turku, Sanna Pätsi).

detailed description of the sampling principle can be found in Bohlmann et al. (2019) and Hirst (1952).

During our campaign, 18 different pollen types have been identified with the Hirst-type air sampler. The most abundant pollen types are birch (*Betula*), pine (*Pinus*) and spruce (*Picea*), which make up 93 % of the total annual pollen count in 2019. Birch pollen grains are almost spherical, around 20–30 µm in diameter and have three pores on their surface. The surface pattern is rather smooth (Blackmore et al., 2003). Pine and spruce pollen belong to the *Pinaceae* family. Their pollen grains are large and non-spherical. They possess two air bladders which assist those pollen grains to be dispersed by wind despite their large size. The diameter of pine pollen grains on their longest axis is around 65–80 µm, while spruce pollen is considered very large for anemophilous pollen, with a diameter around 90–110 µm on the longest axis (Nilsson et al., 1977). The surface of the pollen corpus of spruce pollen is wrinkled and wavy (Grímsson and Zetter, 2011; Shen et al., 2020). Figure 1 shows examples of micrographs during our measurement period. Examples of birch and spruce pollen grains are marked with green and red arrows, respectively. The different size dimension of birch and spruce pollen is clearly visible.

2.4 In situ measurements

Various in situ aerosol instruments were exploited for characterizing the overall aerosol load in the air at the site. An optical particle sizer (OPS; TSI Inc., model 3330) provided particle number concentration and size distribution in the size range of 0.3–10 µm, which were used to calculate the volume size distribution (by assuming spherical shape for the particles). A Synchronized Hybrid Ambient Real-time Particulate Monitor (SHARP; Thermo Fisher Scientific Inc., model 5030) and an aethalometer (Magee Scientific, model AE-31) provided continuous total particle mass and black carbon (BC) concentration, respectively. The instruments resided in the container and sampled through vertical stainless-steel lines with total air inlets. Meteorological conditions, such as temperature, relative humidity and wind speed and di-

rection, may have effects on pollen concentration and distribution in the atmosphere. At Vehmasmäki, these meteorological parameters are measured at several heights between 2 and 300 m on the co-located mast. The lowest height for wind speed and direction is 26 m, which is around the average canopy height at this site.

3 Results and discussion

During our 4-month campaign in 2019, several periods of high pollen concentrations have been observed. One particular intense event with unusually high depolarization ratios and high pollen concentration will act as an example period for this study. An overview of this intense period is given in Fig. 2. From 16 to 19 May 2019, high bi-hourly concentrations of birch pollen, reaching up to 9000 grains m⁻³, were detected. The share of spruce pollen to the total pollen number concentration increased from 0.2 % on 16 May to 22 % on 19 May. Other pollen types only accounted for 0.8 % of the total pollen concentration during the period (Fig. 2a) and thus can be neglected.

The time–height display of Polly^{XT} and Halo Doppler lidar observations is shown in Fig. 2b–d and e, respectively. During daytime, a high aerosol load was observed within the first 2 km considering the strong range-corrected signal at 1064 nm (Fig. 2b). Also the VDR at 532 nm and the Halo particle depolarization ratio at 1565 nm show strongly enhanced values and are increasing towards 19 May (Fig. 2d, e). At 355 nm, however, the VDR does not show significant features except a faint increase up to 2 km on 19 May. This can be explained by the strong influence of molecular scattering on the VDR at 355 nm and the consequently smaller effect of particles compared to larger wavelengths. Faint elevated layers between 3 and 5 km were observed on all 4 d especially during night. Those layers seem to be weakly depolarizing. Backward trajectories (not shown here) indicate that during all 4 d, the air masses arriving at different heights from 500 m to 5 km were advected from northwest and passed over northern Scandinavia and the Norwegian Sea; i.e., they

Figure 2. Overview of the 16–20 May 2019 period. Pollen concentration detect by Hirst-type spore trap at 4 m height (a), range-corrected signal at 1064 nm (b), linear volume depolarization ratio at 355 (c) and 532 nm (d) by Polly^{XT} Raman polarization lidar, Halo Doppler lidar depolarization ratio at 1565 nm (e) and automatic layer detection (f).

did not pass over dust-influenced or highly polluted areas. Thus, it can be assumed that the studied air masses were not influenced by depolarizing dust. The depolarizing elevated aerosol layers between 3 and 5 km are likely pollen lifted by turbulence or long-range-transported pollen as other sources of depolarizing aerosols are missing along the trajectory.

3.1 Case study with highly depolarizing aerosol

Here, 19 May is of special interest. Lidar measurements show a strongly increased backscatter coefficient at 1064 nm and increased volume depolarization ratios especially at 532 and 1565 nm (Fig. 2). The depolarization ratios at these wavelengths are exceptionally high compared to commonly observed depolarization ratios for non-spherical particles. However, measurements with the same measurement settings prior to the intense pollen period show usual values. We therefore consider our depolarization ratio measurements trustworthy.

In Fig. 3, 19 May is presented separately, showing the range-corrected signal at 1064 nm (Fig. 3a) and the volume depolarization ratio at 532 nm (Fig. 3b). The period from 11:00 to 13:00 UTC (shown within white bars) was selected to present time-averaged lidar profiles (Fig. 3c–e). For processing the lidar measurements, a lidar ratio of 60 sr was applied as it was determined for birch and spruce mixtures in our previous study (Bohlmann et al., 2019). Relative humidity and temperature profiles according to GDAS1 (Global Data Assimilation System) data are shown in Fig. 3f. According to profiles of the backscatter signal and relative humidity, the top of the boundary layer is at around 2.5 km. Above 8 km, cirrus clouds have been detected, which show typical depolarization values for cirrus clouds of around 0.4 at 355 and 532 nm (Chen et al., 2002; Sassen and Zhu, 2009; Voudouri et al., 2020). Within the boundary layer, the backscatter coefficient at 532 nm is larger than at 355 nm, resulting in a negative backscatter-related Ångström expo-

Figure 3. Case study on 19 May 2019. Range-corrected lidar signal at 1064 nm (a) and volume depolarization ratio at 532 nm (b). Time-averaged profiles of backscatter coefficients (c), backscatter-related Ångström exponents (d) and depolarization ratios (e) of the selected time period, 11:00–13:00 UTC (white lines), are given. Profiles of temperature and relative humidity from GDAS1 data at 12:00 UTC (f).

nent (BÅE) at 355/532 nm of -0.64 ± 0.45 . Cases of negative Ångström exponents have previously been observed for dust particles (Bohlmann et al., 2018; Rittmeister et al., 2017; Tsekeri et al., 2017; Veselovskii et al., 2016) and are likely caused by specific chemical compositions causing a spectral variation of the imaginary part of the refractive index (Veselovskii et al., 2016). The BÅE at 532/1064 nm, however, is positive (0.97 ± 0.41) and comparable with values of dust smoke mixtures (Kanitz et al., 2014; Tesche et al., 2011). The negative BÅE at 355/532 nm, while having a positive BÅE at 532/1064 nm, could be characteristic of pollen.

VDR and PDR values within the boundary layer are high. Mean PDRs are 0.06 ± 0.02 , 0.46 ± 0.22 and 0.35 ± 0.07 at 355, 532 and 1565 nm, respectively. Very high depolarization ratios of aerosol particles have been detected with lidar before. Ansmann et al. (2009) measured depolarization ratios of 0.5–1 at 710 nm during the Saharan Mineral Dust Experiment (SAMUM). Those high depolarization ratios were caused by large dust, silt-sized and sand particles ($> 10 \mu\text{m}$) which were lifted by convective plumes and dust devils. In our observations, the depolarization ratios at 355 and 532 nm show a decline with height, while the Halo PDR at 1565 nm is almost constant throughout the whole boundary layer. Values of PDR, as well as BÅE, always represent a mixture of pollen and background aerosol. We assume that the concen-

tration of pollen decreases with height, i.e., increasing distance to the pollen source, and the influence of background aerosol increases. The longer Halo wavelength is more sensitive to larger particles (pollen) and less sensitive to smaller particles (background aerosol), and thus the depolarization ratio does not decrease with height in contrast to the PDR at 355 and 532 nm. The different sensitivity to background aerosol could also explain the difference of the PDR at 355 and 532 nm. Later in this study, in Sect. 3.6, we will use a method described by Shang et al. (2020) to separate the influence of background and the pollen on measured optical properties.

3.2 Case studies with different pollen concentrations

Figure 4 shows profiles of the Polly^{XT} backscatter coefficients and linear particle depolarization ratio at 355 and 532 nm as well as the attenuated backscatter and PDR at 1565 nm by the Halo Doppler lidar for a selected 2 h time period on each day representing different pollen conditions. The bar chart illustrates the pollen concentration obtained from the Hirst-type pollen trap during the time period each day. Green color represents the birch pollen concentration, while red represents the spruce pollen concentration. On 19 May, the total pollen concentration and the share of spruce pollen were the highest. On 16 and 17 May, however, no spruce pollen was detected. A strong backscattering signal was detected up to 2.5 km on 19 May, whereas on the other days the signal was weaker, and according to the strong decrease of the backscatter signal, the boundary layer height was lower. Most noticeable is the increase of the PDR at 532 and 1565 nm on the day with the highest spruce pollen share and total pollen concentration, while the PDR at 355 nm is around the same on all days and clearly lower than at the longer wavelengths.

The increased depolarization on 19 May is likely related to the large amount of highly non-spherical and large spruce pollen on this day compared to the other days. In Fig. 1, micrographs of the pollen observations on 18 and 19 May from 09:00 to 11:00 UTC are shown. Birch and spruce pollen was observed on both days, but the amount of both birch and spruce pollen was clearly higher on 19 May. This may have resulted in the higher depolarization observed on this day. Certainly, other factors, such as ambient weather conditions or the fragmentation of pollen grains, could have contributed to this observation as well. Those effects will be investigated in the following section.

3.3 Findings from supporting data

Pollen release and distribution is heavily influenced by the ambient weather conditions. Higher temperature and wind speed positively affect the pollen concentration, while the relative humidity and pollen concentration are negative related (Bartková-Ščevková, 2003; Noh et al., 2013). Under

Figure 4. Case studies from each day between 09:00 and 11:00 UTC. Backscatter coefficient and particle depolarization ratio at 355 and 532 nm by Polly^{XT} (a–d) and the attenuated backscatter and particle depolarization ratio at 1565 nm by Halo (e–f). The bar chart shows the pollen concentration during the 2 h period each day, with green representing birch and red representing spruce pollen.

certain meteorological conditions, e.g., humidity changes and turbulent disturbances, pollen grains can rupture to smaller fragments (Hughes et al., 2020; Miguel et al., 2006; Taylor et al., 2002, 2004). The pollen fragments are not recorded as pollen or counted and therefore do not contribute to the pollen count using microscopic analysis. However, the fragments can persist much longer in the atmosphere than intact pollen due to their smaller size and lower falling velocity. The small pollen fragments can act efficiently as ice-nucleating particles (von Blohn et al., 2005; Diehl et al., 2001, 2002) and penetrate deeper into the respiratory system than intact pollen where they can trigger severe allergic reactions and asthma. Furthermore, small, non-spherical pollen fragments could cause a higher depolarization ratio.

Figure 5 gives an overview of the meteorological conditions during the studied period. The temperature and relative humidity at 2 m (Fig. 5b) and wind speed, wind direction and gust wind at 26 m (Fig. 5c) above ground are shown in addition to the pollen concentration (Fig. 5a). The diurnal cycles of temperature and relative humidity are clearly visible with a maximal relative humidity during the night and maximal temperature shortly after noon. Wind measurements show only a weak diurnal change with a slightly lower wind speed in the evening hours. On 19 May, the wind speed increased rapidly before noon, and a more distinct diurnal cycle compared to the other days is apparent. At the same time, the temperature was higher and the relative humidity slightly lower than on the preceding days, which could explain the higher pollen concentration on this day. The local wind direction changed from northwest on 16 May to southeast on 19 May. Gust winds predominately follow the trend of the mean wind speed, although on 17 May, the stronger gust winds at around 19:00 UTC could explain the high amount of birch pollen at this time.

Turbulent movements within the boundary layer can affect the dispersal of airborne pollen, and pollen grains can be lifted vertically to heights where they are detectable with lidar. Moreover, turbulent conditions could cause pollen grains to rupture upon impact with solid surfaces or other aerosols (Visez et al., 2015). The TKE dissipation rate obtained from the Halo Doppler lidar is shown in Fig. 5d. However, no significant difference between the 4 d was found to indicate a higher occurrence of pollen fragments and further explain the higher depolarization ratio on the last day. The higher detection height on 19 May is caused by a higher aerosol load and therefore higher signal-to-noise ratio on that day.

In situ measurements have been checked to characterize the ground-level aerosol conditions at our measurement site. The particle number concentration at ground level was higher on 18 and 19 May than on 16 and 17 May (Fig. 6a). The evolution of the volume size distribution (Fig. 6b) shows an increase in the concentration of 3–10 μm particles, which could be fragments of birch or spruce pollen. The increase in the coarse particle fraction was verified with the total mass concentration (data not shown). The BC concentration (not shown) was low (around 0.1–0.2 $\mu\text{g m}^{-3}$) throughout the period, except for a short peak on 18 May at 18:00 UTC (also visible in the OPS data; Fig. 6a); thus, no smoke contribution in the boundary layer is expected.

Our results indicate that the influence of pollen fragments is uncertain as the amount of pollen fragments is not recorded by the Hirst-type pollen sampler, and in situ, meteorological and Halo turbulence measurements do not suggest apparent reasons for a pronounced fragmentation of pollen grains in the atmosphere. However, pollen fragments could cause depolarization in the lidar signal and also partly explain the wavelength dependence of the PDR. Therefore, the impact of pollen fragments needs to be explored in future studies.

Figure 5. Overview of pollen concentration by Hirst-type spore trap (a), air temperature and relative humidity at 2 m mast height (b), wind speed, wind direction and wind gust at 26 m mast height including wind direction shown by color (c) and turbulent kinetic energy (TKE) dissipation rate (d) by Halo Doppler lidar.

Figure 6. Time series of particle number concentration (a) and volume size distribution in the range 0.3–10 μm (b) by optical particle sizer (OPS) using a 10 min time average.

3.4 Lidar-derived optical parameters

To investigate the relationship between lidar-derived optical parameters and the pollen observations and to define characteristic values for specific pollen conditions, we determined mean values in the detected layers shown in Fig. 2f. We here focus on the lowest detected layer as it can be assumed to contain most of the local pollen concentration. A total of 47 pollen layers has been detected in the 2 h averaged dataset of the 4 d period. Mean values of the lidar-derived optical properties are presented in Figs. 7 and 8. Color represents the share of spruce pollen of the total pollen number concentration detected by the Hirst-type spore trap. Size of the markers

represents the total 2 h pollen concentration. Daily mean values are shown with black markers; error bars represent the standard deviation. The lower limit for reliable profiles was about 350 m after combining near- and far-field channels and vertical smoothing, whereas a higher layer bottom limit of 900 m was applied on the profiles of PDR at 355 nm (Fig. 7a) and the Ångström exponent at 532/1064 nm (Fig. 8b) due to the overlap problems at 355 nm and the higher full overlap height at 1064 nm, respectively.

In Fig. 7, the particle depolarization ratios at all three wavelengths are compared. Two groups depending on the spruce pollen share are apparent in the comparison of PDR at 532 against 355 nm (Fig. 7a) and PDR at 532 against 1565 nm (Fig. 7b). Cases with a high share of spruce pollen and high total pollen concentration tend to show the highest PDR values ranging between 0.4 and 0.9 at 532 nm, while the PDR for cases with a lower share of spruce pollen is below 0.4. The PDR at 355 nm is below 0.17 for all cases, and no correlation with the spruce pollen share is apparent. The relationship between the PDR at 1565 and 532 nm is nearly linear when the spruce share is low (Fig. 7b). With increasing spruce pollen share, the PDR at 532 nm exceeds the PDR at 1565 nm. This wavelength dependency of the PDR with increasing spruce pollen share could be characteristic of this large and non-spherical pollen type and provide a means for classifying conifer pollen.

Figure 8 shows the backscatter-related Ångström exponents at 355/532 nm and 532/1064 nm against the PDR at

Figure 7. Particle depolarization ratio (PDR) as measured with Polly^{XT} at 355 vs. the PDR at 532 nm (a) and Halo PDR at 1565 nm vs. Polly PDR at 532 nm (b). Color represents the share of the spruce pollen concentration to the total pollen count; size denotes the total pollen concentration. Black markers present daily mean values and their standard deviation.

532 nm. The tendency towards smaller Ångström exponents with increasing depolarization ratio indicates the increasing impact of larger and non-spherical particles such as spruce pollen. Similar behavior has already been reported in Bohlmann et al. (2019) and Shang et al. (2020). For cases with a high amount of large, non-spherical spruce pollen, e.g., with > 15 % share of the total pollen number concentration, the BÅE at 355/532 nm is negative. Previously, negative Ångström exponents have only been reported for dust close to the dust source region where very large dust particles are present (Hofer et al., 2020; Rittmeister et al., 2017; Veselovskii et al., 2016). This feature, however, could also be characteristic of aerosol conditions with the presence of very large pollen grains.

Table 1 summarizes the daily mean values shown in Figs. 7 and 8 with black markers. Mean values are given for different layer bottom heights as profiles could only be retrieved down to 900 m at 355 and 1064 nm. Noteworthy is the spectral dependence of the depolarization ratio, which is also illustrated in Fig. 9.

The daily mean backscatter-related Ångström exponent is clearly different on the spruce-dominated day. While it is around 0.9–1.3 at 355/532 nm and 1.18–1.52 at 532/1064 nm on the days with low spruce pollen share, BÅEs are clearly smaller at both wavelength pairs on the spruce-dominated day, suggesting the presence of larger particles. The nega-

Figure 8. Backscatter-related Ångström exponent (BÅE) at 355/532 nm (a) and 532/1064 nm (b) vs. the PDR at 532 nm. Color represents the share of the spruce pollen concentration to the total pollen count; size denotes the total pollen concentration. Black markers present daily mean values and their standard deviation.

tive BÅE at 355/532 nm might be characteristic of large, non-spherical spruce pollen grains since it has only been observed on days with a high spruce pollen concentration. It must be noted that the Ångström exponent also depends on the background aerosols and its use to characterize pollen needs to be considered carefully. However, under the same background conditions, the Ångström exponent can be an indicator for the pollen type. As the considered time period is rather short and no far-range transported aerosol sources were observed, the background conditions can be assumed stable during the period. The change of the Ångström exponent can therefore be attributed to the presence of pollen.

3.5 Spectral dependence of the linear depolarization ratio

Figure 9 shows the daily mean linear particle depolarization ratios detected with Polly^{XT} (at 355 and 532 nm) and the Halo Doppler lidar (at 1565 nm). The PDR at 355 nm is around 0.1 on all 4 d. Of special interest is the strong spectral dependency of the PDR observed on 19 May. A higher depolarization ratio was observed at 532 than at 1565 nm. On the preceding days, however, the depolarization ratios at 532 and 1565 nm are only slightly higher than at 355 nm and almost equal. As shown in Table 1, the amount of observed spruce pollen was increasing each day and contributed to almost 22 % of the total number concentration on 19 May. Longer

Table 1. Daily mean values of pollen concentrations and lidar-derived optical parameters of the pollen layers. The total pollen (spruce pollen) concentrations are measured by the Hirst-type volumetric air sampler. Layer heights are determined using the gradient method. Daily mean values and standard deviation of lidar-derived optical parameters in the pollen layers using a layer bottom height of 900 m (a) and 350 m (b).

Date in 2019 (dd.mm)	16.05	17.05	18.05	19.05
Daily mean total (spruce) pollen concentration [m^{-3}]	3653 (8)	3157 (63)	1727 (132)	3553 (776)
Layer top height (km)	1.47 ± 0.42	1.41 ± 0.23	1.01 ± 0.08	1.23 ± 0.41
(a)				
PDR 355	0.08 ± 0.03	0.09 ± 0.03	0.11 ± 0.03	0.10 ± 0.02
PDR 532	0.10 ± 0.03	0.16 ± 0.10	0.15 ± 0.07	0.38 ± 0.23
PDR 1565	0.07 ± 0.06	0.16 ± 0.14	0.16 ± 0.12	0.29 ± 0.10
BÅE 355/532	1.19 ± 0.41	0.92 ± 0.39	1.22 ± 0.80	-0.18 ± 0.96
BÅE 532/1064	1.20 ± 0.29	1.18 ± 0.74	1.52 ± 0.49	1.08 ± 0.46
Backscatter coefficient 355 [$\text{Mm}^{-1} \text{sr}^{-1}$]	0.44 ± 0.15	0.38 ± 0.09	0.48 ± 0.14	0.99 ± 0.47
Backscatter coefficient 532 [$\text{Mm}^{-1} \text{sr}^{-1}$]	0.27 ± 0.08	0.26 ± 0.06	0.33 ± 0.17	1.27 ± 0.86
(b)				
PDR 532	0.14 ± 0.03	0.20 ± 0.11	0.22 ± 0.11	0.46 ± 0.26
PDR 1565	0.13 ± 0.06	0.19 ± 0.10	0.21 ± 0.10	0.30 ± 0.09
BÅE 355/532	1.29 ± 0.52	0.92 ± 0.36	0.90 ± 0.68	-0.10 ± 0.92
Backscatter coefficient 355 [$\text{Mm}^{-1} \text{sr}^{-1}$]	0.44 ± 0.12	0.40 ± 0.06	0.62 ± 0.16	1.01 ± 0.44
Backscatter coefficient 532 [$\text{Mm}^{-1} \text{sr}^{-1}$]	0.26 ± 0.07	0.28 ± 0.05	0.47 ± 0.23	1.26 ± 0.85

Figure 9. Spectral dependence of the mean particle depolarization ratio for each day. Additionally, mean linear depolarization ratio of pine and birch pollen determined in laboratory measurements by Cao et al. (2010) are shown. Error bars represent the standard deviation.

wavelengths are more sensitive to larger particles and less influenced by Rayleigh scattering (Ansmann et al., 2009). The strong spectral dependency on 19 May can therefore be explained by the high amount of non-spherical spruce pollen on that day. Another explanation for the observed wavelength dependence could be the surface structure of pollen. Patterns on the outer wall of the pollen, the exine, can vary enormously in their appearance, but within the same species patterns are usually similar (Wang and Dobritsa, 2018). The

non-smooth pattern on spruce pollen grains could additionally cause a depolarization of the backscattered light.

A spectral dependence of the linear depolarization ratio of pollen has already been reported by Cao et al. (2010), who measured the depolarization ratio for different pollen types, such as birch and pine, in controlled laboratory experiments. For birch pollen, Cao et al. (2010) reported a PDR of 0.08 ± 0.01 , 0.33 ± 0.00 and 0.30 ± 0.01 at 355, 532 and 1570 nm, respectively. No measurements have been conducted for spruce pollen. However, the authors state values for pine which belong to the same family as spruce and have smaller but in shape comparable pollen grains. According to Cao et al. (2010), depolarization ratios for pine pollen are 0.20 ± 0.01 , 0.41 ± 0.01 and 0.28 ± 0.01 at 355, 532 and 1570 nm, respectively. The laboratory-derived depolarization ratios by Cao et al. (2010) are shown for comparison in Fig. 9. Empty markers represent the lidar depolarization ratios for paper birch (*Betula papyrifera*) and Virginia pine (*Pinus virginiana*). Especially the spectral dependence for pine pollen is very similar to our observations on the spruce-dominated day, having the peak at 532 nm. It should be kept in mind that the laboratory measurements were performed in an aerosol chamber and that they represent the depolarization ratio of the pure and dry pollen. Furthermore, pollen from different species was used compared to what we detected at our measurement site. Different values were therefore expected. However, the fact that we detected similar wavelength dependence in ambient conditions to that in the laboratory measurements suggests that the wavelength

Figure 10. Frequency distribution of backscatter-related Ångström exponent at 355/532 nm of the pollen layers detected during the studied period. The related η value, as used in Shang et al. (2020), is given on the second x axis. The BÅE threshold of -0.4 to remove “extreme cases” is marked by the dashed vertical line.

dependence with maxima at 532 nm might be characteristic of pollen especially for large and non-spherical conifer pollen types.

The spectral dependence of the PDR has previously been reported for different aerosol types. Haarig et al. (2018) and Hu et al. (2019) observed a completely different spectral dependence of the PDR for stratospheric smoke. The PDR is strongly decreasing with increasing wavelength caused by the dominance of small particles ($< 0.5 \mu\text{m}$). A similar spectral dependence of the PDR with the peak PDR at 532 nm and decreasing towards 355 and 1064 nm, however, was reported by Haarig et al. (2017) and Burton et al. (2015) for transported Saharan dust. As for pollen, the optical properties of dust are dominated by large particles ($> 1 \mu\text{m}$) as well. However, depolarization ratios at 355 nm are clearly smaller for pollen than for dust, while the PDR at longer wavelengths is higher, which allows the distinction of pollen and aged dust.

3.6 Estimation of the pollen depolarization ratio values

To estimate the depolarization ratio values for pure pollen, a newly developed method by Shang et al. (2020) was applied to the lidar measurements. Mathematically, there is a power-law relationship between the total backscatter-related Ångström exponent and the pollen backscatter contribution (the ratio of pollen backscatter coefficient and the total particle backscatter coefficient). The depolarization ratio at 532 nm of pure pollen was investigated using the BÅE at 355/532 nm. Our results of this intense pollen event show two groups of data depending on the spruce pollen share: (i) a quite good power-law relationship is found for cases with a low share of spruce pollen; (ii) the BÅE at 355/532 nm is almost constant with increasing PDR for cases with a higher share of spruce pollen. These two groups can also be identified as the two modes in the frequency distribution of BÅE values at 355/532 nm presented in Fig. 10. The first mode of

Figure 11. Assumed pollen backscatter-related Ångström exponent at 355/532 nm against the related pollen depolarization ratio at 532 nm for the studied period after removing cases below the BÅE threshold of -0.4 . The pollen depolarization ratio is the calculated depolarization ratio value for a pollen backscatter contribution of 1 at assumed pollen BÅE.

values around -0.9 , denoted as “extreme cases”, is related to PDR larger than 0.25, mostly for the data on the last day (high spruce share). For these cases, the BÅE is almost constant throughout the boundary layer, whereas the depolarization ratio is decreasing with height. This reveals the limitation of Ångström exponent at lidar wavelengths of 355 and 532 nm for the detection of big pollen particles since the BÅE normally decreases with increasing share of pollen particles, i.e., increasing PDR.

A threshold of -0.4 was applied to remove those “extreme cases” before applying the algorithm of Shang et al. (2020). A depolarization ratio of 0.03 at both 355 and 532 nm was used for non-pollen particles, which is a mean value for pollen-free periods at our measurement site. After the iteration, we found the relationship between the pollen BÅE at 355/532 nm and the pollen depolarization ratio at 532 nm presented in Fig. 11. This relationship is representing the mixture of birch and spruce pollen, with low spruce pollen share after the selection of the threshold. Our previous studies (Bohlmann et al., 2019; Shang et al., 2020) indicate that pollen consists of medium- to high-absorbing particles with lidar ratio values from 55 to 70 sr at both 355 and 532 nm for birch and spruce pollen, without significant wavelength dependence. The pollen grains are quite big and thus can be assumed to be wavelength independent on the backscatter at wavelengths of 355 and 532 nm. Under the assumption that the backscatter-related Ångström exponent at 355/532 nm should be zero for pollen, the pollen depolarization ratio was found to be 0.44 for the mixture of birch and spruce pollen observed during this intensive period of high pollen concentrations. The spruce pollen share to the total pollen number

concentration of the birch–spruce pollen mixture ranges between 0 % and 35 % for the estimated pollen depolarization value. The depolarization at 355 nm was calculated using the pollen backscatter contribution at 355 nm and the BÅE at 355/532 nm. Assuming a BÅE of zero, a pollen depolarization ratio of 0.16 at 355 nm was found for the birch and spruce mixture. In order to retrieve the pollen depolarization ratio of the “extreme cases”, we used the BÅE at 532/1064 nm, as longer lidar wavelengths might be beneficial for cases with a high share of very large particles. However, our dataset of corresponding cases is too limited to lead to a meaningful conclusion. More cases with high spruce concentration are needed.

4 Conclusion

During the pollen campaign in 2019 at the European Aerosol Research Lidar Network (EARLINET) station in Vehmasmäki (Kuopio), Finland, we were able to expand the dataset of birch and spruce pollen observations with improved lidar-based measurements down to around 350 m. In previous studies, measurements were restricted by a high overlap height which limited usable profiles to data above 800 m. The majority of strongly depolarizing spruce pollen, however, is assumed to be close to the pollen source near ground level. Thus, when conducting pollen measurements with lidar, the lower limit of usable lidar data should be as low as possible. In our study, we utilized depolarization measurements of a co-located Halo Doppler lidar to add a third depolarization wavelength. Very high depolarization ratios have been detected during a 4 d period of birch and spruce pollen with an increasing amount of spruce pollen towards the end. In situ aerosol measurements and meteorological data have been considered to characterize the overall aerosol load in the air and the air masses at the site. Ruptured pollen could affect the observed depolarization ratio, as the smaller fragments of ruptured pollen are highly non-spherical. We detected a slightly higher number of particles in the diameter range from 3–10 µm on the day with high depolarization, which could be partly caused by pollen fragments. However, more research on the impact of pollen fragments is needed and the amount of pollen fragments should be recorded in future studies. Also the possible impact of the pollen surface structure on the measured depolarization ratios needs to be investigated, as the rough surface of certain pollen types can cause higher depolarization ratios.

The investigation of lidar-retrieved optical properties in detected pollen layers revealed a wavelength dependence of the depolarization ratio especially in the presence of spruce pollen. On a day with a spruce pollen share of the total pollen number concentration of about 22 %, high PDR values of 0.46 ± 0.26 and 0.30 ± 0.09 were detected at 532 and 1565 nm, respectively, in pollen layers down to around 350 m. The PDR at 355 nm could only be detected down to

900 m. In those layers, mean PDR values were 0.10 ± 0.02 , 0.38 ± 0.23 and 0.29 ± 0.10 at 355, 532 and 1565 nm, respectively. The wavelength dependency could be explained with the higher sensitivity of the longer lidar wavelengths to big pollen particles ($\sim 100 \mu\text{m}$) and characteristic surface pattern of such pollen. This wavelength dependence could be characteristic of large, non-spherical spruce pollen. Furthermore, negative backscatter-related Ångström exponents have been detected when the spruce pollen share was high. Negative Ångström exponents have previously been detected for dust but could also be a characteristic feature of spruce pollen. A novel method introduced by Shang et al. (2020) was applied to the measurement data, and pollen depolarization ratio values of 0.44 at 532 nm and 0.16 at 355 nm were determined for the birch–spruce pollen mixture. Furthermore, a limitation of the Ångström exponent at the lidar wavelengths of 355/532 nm for the characterization of very large pollen particles like spruce was found.

To determine values of pure pollen, more cases with high concentrations of only one pollen type are necessary. We found characteristic values for atmospheric conditions in the presence of spruce pollen, but at our measurement site spruce pollen almost always occurred simultaneously with other pollen types. This complicates the investigation of pollen type characteristics, as optical properties vary depending on different mixtures of backscattering aerosol. Furthermore, pollen fragments and the pollen surface structure need to be considered when investigating pollen properties with lidar because non-spherical fragments and pollen-type-specific surface structures possibly affect the detected depolarization ratio as well. Nevertheless, our study shows the spectral dependence of the depolarization ratio for pollen, especially spruce, in the atmosphere.

Data availability. Lidar data are available upon request from the authors and data “quicklooks” are available on the PollyNET website (<http://picasso.tropos.de/>, last access: 17 December 2020, PollyNET, 2020).

Author contributions. SB analyzed the data and wrote the manuscript in close cooperation with XS. VV provided the Halo Doppler lidar data. AL provided and assured the quality of the in situ measurements. SP analyzed the pollen samples. MK and EG initiated the project. All authors contributed to the scientific discussion and the article preparation.

Competing interests. The authors declare that they have no conflict of interest.

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