Long-term aerosol particle depolarization ratio measurements with Halo Doppler lidar

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Abstract. It has been demonstrated that Halo Doppler lidars have the capability for retrieving the aerosol particle ~~linear~~ depolarization ratio at a wavelength of 1565 nm. For these lidars operating at such a long wavelength, the retrieval quality depends to a large degree on an accurate representation of the instrumental noise floor and the performance of the internal polarizer, whose stability have not yet been assessed in long-term operation~~However, the retrieval depends on an accurate representation of the instrumental noise floor and the performance of the internal polarizer, whose stability have not been assessed in long-term operation~~. Here, we use four years of measurements at four sites in Finland to investigate the long-term performance of Halo Doppler lidars focusing on ~~for~~ aerosol particle depolarization ratio retrieval. The instrumental noise level, represented by noise-only signals in aerosol- and hydrometeor- free regions, shows stable performance for most instruments, but clear differences between individual instruments. For all instruments, the polarizer bleed-through evaluated at liquid cloud base remains reasonably constant at approximately 1% with a standard deviation less than 1%. We find these results sufficient for long-term aerosol particle ~~linear~~ depolarization ratio measurements and proceed to analyse the seasonal and diurnal cycles of the aerosol particle depolarization ratio in different environments in Finland including in the Baltic Sea archipelago, boreal forest and rural sub-arctic. To do so, we further develop the background correction method and construct an algorithm to distinguish aerosol particles from hydrometeors. The four-year averaged aerosol particle depolarization ratio ranges from 0.07 in sub-arctic Sodankylä to 0.13 in the boreal forest in Hyytiälä. At all sites, the aerosol particle depolarization ratio is found to peak during spring and early summer, even exceeding 0.20 at the monthly-mean level, which we attribute to a substantial contribution from pollen. Overall, our observations support the long-term usage of Halo Doppler lidar depolarization ratio including detection of aerosols that may pose a safety risk for aviation.

1 Introduction

Information on the aerosol vertical distribution in the atmosphere is vital for many applications. For instance, the direct radiative effects of aerosols can be quite different if the aerosol layer is situated above a cloud layer rather than within the boundary layer, while aerosol indirect radiative effects occur only if aerosols are immersed within the cloud (IPCC, 2021). The impact of aerosol on clouds and the radiative balance of the Earth are among the largest uncertainties in our understanding (IPCC, 2021). Air quality and associated adverse health effects (Di et al., 2017) are determined by surface concentrations, which, however, are strongly affected by the vertical structure of the boundary layer (Kanawade et al., 2020; Wang et al., 2020). Finally, from an aviation point of view, high resolution profiles of aerosol vertical distribution could play a crucial role in mitigating the impact of hazardous aerosol emissions (Hirtl et al., 2020).

Aerosol vertical profiles can be observed with a number of different methods, such as in-situ instruments mounted on different platforms including research and commercial aircraft (Johnson et al., 2008; Pratt and Prather, 2010), tethered balloons (Rankin and Wolff, 2002; Hara et al., 2013; Creamean et al., 2021), hot air balloons (Petäjä et al., 2012), zeppelin (Rosati et al., 2016) and unmanned aerial vehicles (Brus et al., 2021; Mamali et al., 2018). Comprehensive aerosol properties can be obtained from in-situ measurements: mass, size distribution, chemical composition; but the capabilities of the chosen platform limit the

temporal resolution at which profiles can be obtained and the vertical extent of the profiling. On the other hand, active remote sensing with lidar only retrieves the optical properties of aerosol but is capable of continuous observations of the vertical structure of the atmosphere. Space-borne lidars such as CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation; Winker et al., 2009) cover the globe but with low temporal and spatial resolution due to their very narrow swath ~~(Baars et al., 2017)~~. Airborne lidars, such as HSRL-1 (High Spectral Resolution Lidar) built and operated by NASA Langley Research Center (Hair et al., 2008), provide good spatial coverage but are costly and only able to operate for a relatively short period at a time. A combination of temporal and spatial coverage can be achieved through ground-based networks of lidars, such as EARLINET (European Aerosol Research Lidar Network; Pappalardo et al., 2014) and Finland's ground-based remote-sensing network (Hirsikko et al., 2014). These lidar networks enable the monitoring in real-time of the aerosol vertical profiles in different environments across a large aera. Consequently, they facilitate the detection of elevated aerosol layers and the investigation of long-term vertical atmospheric properties. ~~These lidar networks enable the monitoring in real-time of vertical profiles of aerosol in different environments, and consequently facilitate the detection of elevated aerosol layers and the investigation of vertical atmospheric properties throughout the seasons.~~

In active remote sensing, one of the most important parameters for characterising aerosol is the ~~aerosol~~ particle depolarization ratio (denoted as δ), ~~($\delta_{aerosol}$),~~ which is the ratio of the co-polar and cross-polar signals backscattered from aerosol. This parameter is used to distinguish between spherical and non-spherical particles (Burton et al., 2012; Mamouri and Ansmann, 2016; Baars et al., 2017) and is therefore essential in differentiating aerosol types (Illingworth et al., 2015) such as smoke, dust, marine and ash. Typically, the δ of aerosol is measured at shorter wavelengths such as at 355 nm, 523 nm, 532 nm, 694 nm, 710 nm or 1064 nm (Murayama et al., 2001; Sassen, 2002; Engelmann et al., 2016; Baars et al., 2016)~~Typically, $\delta_{aerosol}$ is measured using Raman lidar (Engelmann et al., 2016; Baars et al., 2016) or micropulse lidar (Flynn, et al., 2007; Córdoba-Jabonero et al., 2018) at wavelengths such as 355 nm, 532 nm, 710 nm or 1064 nm~~. This study is conducted using data from Halo Doppler lidars at 1565 nm, which is a relatively new addition to the suite of wavelengths used for the δ of aerosol ~~$\delta_{aerosol}$~~ retrieval (Vakkari et al., 2021).

Halo Doppler lidars (Pearson et al., 2009) are the core remote sensing instruments in the Finnish ground-based remote sensing network. In this study, we analysed data from these instruments at four different locations in the network from 2016 to 2019. Liquid clouds were identified and the δ at liquid cloud base was collected to derive the depolarisation bleed-through ~~Liquid clouds were identified and the depolarisation ratio, δ, at liquid cloud base collected to derive the depolarisation bleed through~~ (Vakkari et al., 2021) and its temporal evolution for five different Halo Doppler lidar instruments. In addition, the stability of every Halo Doppler lidar in the network was also assessed through the time series of the signal-to-noise ratio in aerosol- and hydrometeor- free regions. Furthermore, an Aerosol Identification algorithm was created to enable the separation of aerosol and hydrometeors similar to the Cloudnet algorithm ~~an aerosol identification algorithm was created to enable the separation of aerosol and hydrometeors similar to the Cloudnet algorithm~~ (Illingworth et al., 2007; Tukiainen et al., 2020), but based only on one instrument. The algorithm was used to extract the δ in aerosol region (δ of aerosol) ~~$\delta_{aerosol}$~~ at all locations and subsequently, the overall statistics of the δ of aerosol ~~of $\delta_{aerosol}$~~ at 1565 nm in Finland across four years. These statistics can improve aviation safety by providing a baseline of the δ of aerosol ~~$\delta_{aerosol}$~~ at this wavelength, so that potentially hazardous layers such as smoke and volcanic ash can be separated from the natural aerosol more easily.

2 Materials and methods

2.1 Halo Doppler lidar

The Finnish remote sensing network deploys Halo Doppler lidars in several measurement stations across Finland (Hirsikko et al., 2014). This study uses data from Utö, Hyytiälä, Vehmasmäki and Sodankylä (Fig.~~ure~~ 1). Each location has a different

80 environment, enabling comparisons between both urban and rural; marine, continental, and sub-arctic regions. Detailed descriptions and the study period for each location are shown in Table 1.
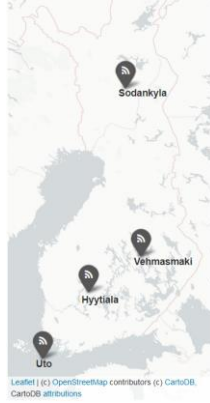

Figure 1: Locations of the instruments across Finland

Site	Description	Instrument	Instrument ID	Study period
Utö 59.77ºN,21.37ºE	Island	StreamLine	Utö-32	1.1.2016 - 16.8.2017
		StreamLine XR	Utö-32XR	22.11.2017 - 31.12.2019
Vehmasmäki - Kuopio 62.89ºN,27.63ºE	Semi-urban/rural	StreamLine Pro	Vehmasmäki-53	1.1.2016 - 31.12.2019
Hyytiälä (SMEAR II) 61.84ºN,24.29ºE	Rural (boreal forest)	StreamLine	Hyytiälä-33	1.1.2016 -7.8.2017
		StreamLine	Hyytiälä-46	9.10.2017 - 31.12.2019
Sodankylä 67.37ºN,26.62ºE	Arctic rural	StreamLine Pro	Sodankylä-54	19.6.2017 - 20.11.2019

85 **Table 1: Description of instrument locations**

The Halo Photonics StreamLine Doppler lidars (Halo Doppler lidars) operated by the Finnish Meteorological Institute are 1565 nm pulsed Doppler lidars transmitting linearly polarized light and equipped with heterodyne detectors that can switch between recording the return in two channels parallel and orthogonal with respect to the transmitted polarisation (Pearson et al., 2009), termed co-polar (parallel) and cross-polar (orthogonal). These lidars are fibre optic systems, utilizing solid-state

90 lasers, and capable of operating continuously for a long period of time (Harvey et al., 2013). In addition, these systems conform to eye-safety requirements as they operate at high-pulse repetition and low-pulse energy mode with a 1565 nm laser (Pearson et al., 2009).

Three versions of Halo Doppler lidars are utilized in this study: StreamLine, StreamLine Pro and StreamLine XR lidars (HALO PHOTONICS | StreamLine series - Product, 2021). The StreamLine and StreamLine XR lidars are capable of full hemispheric

95 scanning. Designed for harsher environments, the StreamLine Pro lidar has no external moving parts, limiting the scanning to within a 20-degree cone around Zenith. The StreamLine XR has higher power and a lower pulse repetition frequency, thus can observe up to 12 km in range above ground level (a.g.l) compared to only 9.6 km for the StreamLine and Streamline Pro. Key specifications of all instruments are shown in Table 2.

The operational mode of each instrument varies with location. The standard operation mode consists of continuous vertical

100 staring with periodical switching to velocity-azimuth display scans for obtaining vertical profiles of the horizontal wind. For

about 10-30 seconds in every hour the instruments perform a periodical background noise determination. Only data from the vertical staring mode is utilized in this study, so occasional gaps in data availability are due to lower elevation angle scanning and the background noise determination.

In vertical staring mode, the instrument emits pulses of polarized light into the atmosphere and then records the returned
105 vertical Doppler velocity (w) and signal-to-noise ratio in co- or cross- polar receiver state (SNR_{co} and SNR_{cross}); receiver is changed between co- and cross- polar sequentially. For a coherent Doppler lidar, attenuated backscatter (β') is calculated from SNR through

$$\beta'(z) = A\frac{SNR(z)}{T_f(z)}, \tag{1}$$

where A incorporates system-specific constants and $T_f(z)$ is the telescope focus function, which depends on range (z), the
110 effective beam diameter and focal length of the system (Frehlich and Kavaya, 1991; Pentikäinen et al., 2020). For the Utö-32XR instrument, data from a co-located Vaisala CL31 ceilometer was utilised to determine the telescope function according to Pentikäinen et al., (2020) and then to calculate β' according to Eq. 1. For the non-XR instruments, β' was determined from the post-processed SNR using the 2 km focal length set in the firmware (Table 2).

Since A and $T_f(z)$ are equal for co- and cross-polar measurements and attenuation can be assumed to be equal for both polarities,
115 the depolarization ratio can be retrieved as:

$$\delta = \frac{SNR_{cross}}{SNR_{co}}. \tag{2}$$

At 1565 nm wavelength, the molecular backscatter coefficient is much smaller than at shorter wavelengths, being approx. $1.44\times10^{-8}\,\mathrm{m}^{-1}\mathrm{sr}^{-1}$ at standard pressure and temperature (Bucholtz, 1995). Additionally, atmospheric transmittance is very close to unity in non-hydrometeor regions (Vakkari et al., 2021). Thus, we consider δ to be a fair estimate of the linear particle
120 depolarization ratio without the correction for the molecular contribution (Vakkari et al., 2021). However, δ is sensitive to the performance of the internal polarizer as well as to the accuracy of the instrumental noise floor for both polarities (Vakkari et al., 2021).

In vertical staring mode, the instrument emits pulses of polarized light into the atmosphere and then records the returned vertical Doppler velocity (w) and signal-to-noise ratio in both co- and cross-polar (SNR_{co} and SNR_{cross}) sequentially. From
125 these measurements, profiles of attenuated backscatter (β') and particle depolarization ratio (δ) are derived.

In Halo Doppler lidars, measurements of SNR_{co} and SNR_{cross} are taken sequentially. For example, if the integration time of the instrument is set to 7 seconds, then SNR_{co} is collected for 7 seconds and then SNR_{cross} is collected during the next 7 seconds (Vakkari et al., 2021), and the resulting δ will be presented with 14 seconds time resolution. Such long-time resolution can cause issues especially for cloud measurements, if e.g., cloud base height changes between co- and cross-polar measurements.
130 However, aerosol measurements in low signal conditions require extended integration time to reduce noise. Thus, integration time is always a compromise between high time resolution and low background noise; and care must be taken to ensure that δ is calculated from the same part of the cloud. On the other hand, aerosol is expected to be well-mixed within each aerosol layer so the δ of aerosol $\delta_{aerosol}$ can beis calculated in this study from 1-hour averaged measurements to minimize noise effect on weak signals.
135 Additionally, data from a co-located Vaisala CL31 ceilometer at Utö was utilised to determine effective beam diameter and focal length according to (Pentikäinen et al., 2020) for the Utö 32XR instrument. For the non-XR instruments, attenuated backscatter was determined from the post-processed SNR using the 2 km focal length set in the firmware (Table 2).

Specification	Values
Wavelength	1565 nm
Beam divergence	3.3e-05 rad

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Laser Energy	1e-05 J
Lens diameter	0.06 m
Number of samples per range gate	10
Range resolution	30 m
Pulse length	200 ns
Minimum range	90 m
Pulse repetition frequency	10 kHz (Utö-32XR)
	15 kHz (Other instruments)
Focus	Infinity (Utö-32XR)
	2 km (Other instruments)
Integration time	30 s (2016-01-01 to 2016-07-12, Utö-32)
	5 s (2016-07-13 to 2017-11-21, Utö-32)
	2 s (Utö-32XR)
	6 s (Hyytiälä-33, Hyytiälä-46, Vehmasmäki-53, Sodankylä-54)
Bandwidth	25000 MHz (Vehmasmäki-53, Sodankylä-54)
	50000 MHz (Other instruments)
Nyquist velocity	9.7 m s^{-1} (Vehmasmäki-53, Sodankylä-54)
	19.4 m s^{-1} (Other instruments)

Table 2: Instrument configurations

2.1.1 Instrumental noise floor

140 Typically, every hour, the instrument performs a background check to determine the range-resolved background noise level, which is then used in the firmware to calculate the SNR. The data in this study have been post-processed with the background correction algorithm as described by Vakkari et al., (2019), which removes the bias in SNR, i.e., SNR is centred on 0 when there is no signal. This algorithm enables the evaluation of the temporal evolution of the SNR in the aerosol- and hydrometeor-free zone, which can be used to assess the long-term changes of the noise floor for each instrument in the network. Here, the

145 aerosol- and hydrometeor- free part of the SNR profiles have been manually collected for all the instruments throughout the whole study period.

2.1.2 Instrumental internal polarizer

In Halo Doppler lidars, an internal polarizer is used to measure the co- and cross-polar signals. The design of the Halo Doppler lidars does not facilitate user calibration of the polarizer performance, unlike aerosol research lidars such as PollyXT (Baars

150 et al., 2011). Therefore, δ at liquid cloud base is used to estimate the internal polarizer performance, or bleed-through. It is defined by Vakkari et al., (2021) as the incomplete extinction in the lidar internal polarizer, which the co-polar signal is leaking into the cross-receiver. This results in a systematic bias in the calculated δ from SNR_{co} and SNR_{cross}Therefore, δ at liquid cloud base, which is defined in this study as

$$\delta = \frac{SNR_{cross}}{SNR_{co}}, \tag{1}$$

155 is used to estimate the internal polarizer performance, or bleed- through (Vakkari et al., 2021).

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Single scattering from a spherical droplet does not change the incident polarization state into the 180-degree backward direction (Liou and Schotland, 1971), which results in no return signal at cross polarization. Hence, it is expected that δ at pure liquid cloud base is zero. However, as the laser beam penetrates further into the cloud, the observed δ gradually increases as the multiple-scattering contribution increases (Liou and Schotland, 1971; Hu et al., 2006), which is demonstrated in ~~figure~~ Fig. 2.

160 In order to determine the long-term performance of the internal polarizer, statistics of δ at liquid cloud base need to be obtained. Previous studies have introduced multiple algorithms based on attenuated backscatter profile to detect liquid cloud base (Zhao et al., 2014; Tuononen et al., 2019). In this study, taking the additional advantage of Halo Doppler lidar's capability in observing δ and w, we develop a new approach based on Tuononen et al., (2019) to detect the liquid cloud base.

First, a period of a liquid cloud layer is chosen through visual inspections. Within this period, the following criteria are used
165 to choose suitable profiles to determine δ at liquid cloud base:

1. $\beta' > 10^{-5}$ m^{-1} sr^{-1},
2. δ increases monotonously from the lowest range gate (cloud base) where β' exceeds 10^{-5} m^{-1} sr^{-1} to the range gate of maximum SNR_{co},
3. vertical extent from the lowest range gate where β' exceeds 10^{-5} m^{-1} sr^{-1} to the range gate of maximum SNR_{co} does
170 not exceed 100 m, and
4. w is between -0.5 m s^{-1} and 0.5 m s^{-1} for the range gates where $\beta' > 10^{-5}$ m^{-1} sr^{-1}.

Criterion 2 reflects the increasing multiple scattering contribution (Liou and Schotland, 1971; Hu et al., 2006). Criterion 3 reflects the rapid attenuation of the signal inside liquid cloud (O'Connor et al., 2004; Tuononen et al., 2019) and reduces the likelihood of including ice clouds in the analysis. Criterion 4 removes profiles that may contain precipitation and to ensure that
175 the observed cloud base does not fluctuate in height too much, i.e., SNR_{co} and SNR_{cross} signals observe the same part of the cloud.

Values of δ at liquid cloud base were collected from each site throughout the whole study period. Where possible, at least one liquid cloud case per week was selected. The mean and standard deviation of δ at all the cloud bases were then calculated and used to determine the bleed-through for each instrument and to investigate its stability over time.

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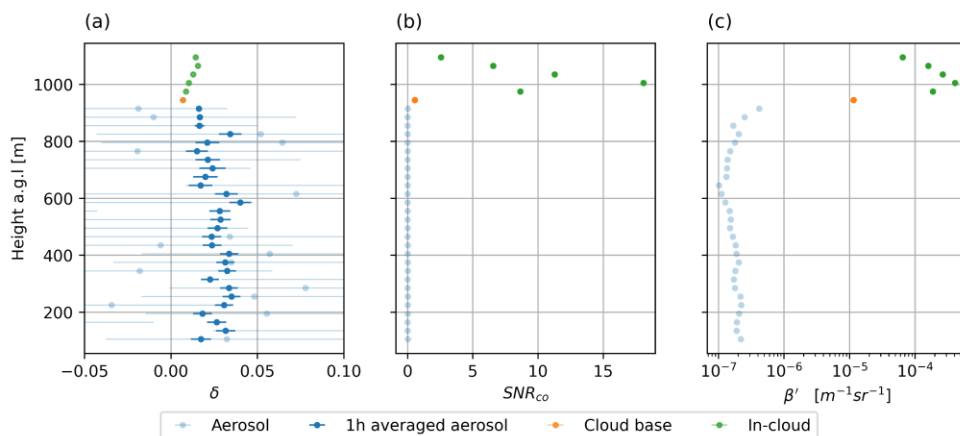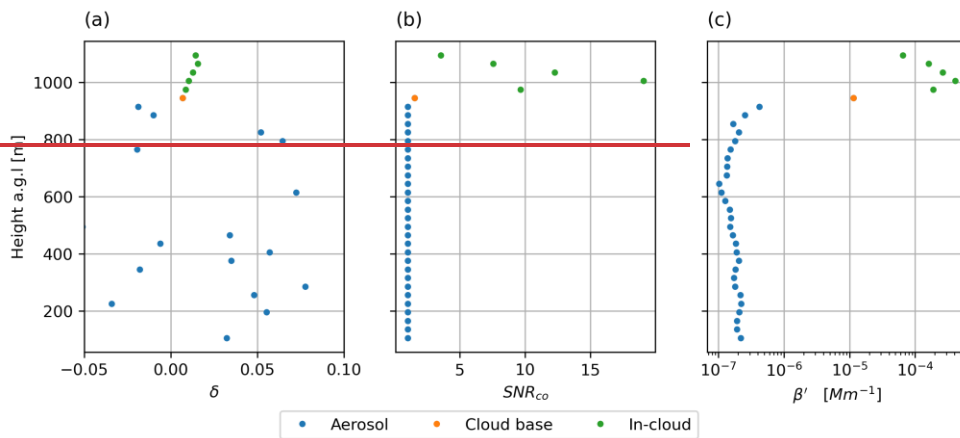
Figure 2: Atmospheric profiles observed on 2018-09-24 at 05:35:17 UTC at Hyytiälä up to 1.4 km a.g.l. a) Depolarization ratio (δ) at the original integration time of 6s and as 1h average for the aerosol part of the profile. Error bar indicates the measurement uncertainty. b) Signal-to-noise ratio in the co-polar channel (SNR_{co}). c) Attenuated backscatter (β').~~Atmospheric profiles observed in 2018-09-24 by Hyytiälä-46 up to 1.4 km a.g.l. a) Depolarization ratio (δ), b) Signal-to-noise ratio in the co-polar channel (SNR_{co}) c) Attenuated backscatter (β')~~

The signal from liquid cloud droplets is very strong, as seen also in Fig.~~ure~~ 2b. Hence, there is a risk of signal saturation. Given the low bleed--through we would expect SNR_{co} to get saturated before SNR_{cross}, which would appear as increasing δ in such cases. Figure S1 displays the 2D histogram of SNR_{co} and SNR_{cross} observed at liquid cloud base in Utö with the XR instrument. SNR_{co} and SNR_{cross} follow a linear relationship with the gradient corresponding to the determined bleed-through, indicating that saturation is not an issue, except maybe for some scattered points where $SNR_{co} > 6$. Depending on cloud base height, the SNR_{co} at cloud base can be as low as 0.01. As seen in Fig S1, there are no non-linear effects in the low SNR_{co} end of the spectrum, and we have no reason to suspect that the polarizer performance would deteriorate with decreasing SNR_{co}. We also note that previous observations with Halo Doppler lidar report near-zero depolarization ratio for marine aerosol, which we expect to be spherical and therefore have very low depolarization ratio (Vakkari et al., 2021). Therefore, we are confident that bleed-through obtained from cloud-base observations can be used to correct also weak aerosol signals.

It should be noted that at the original integration time of 6s, the strong signal from cloud base results in minimal noise level of δ as seen in Fig. 2a. However, for the weak aerosol signal, δ at 6s integration time is rather noisy. Averaging over longer time

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periods reduces the instrumental noise in the aerosol δ substantially, as seen in the 1h averaged δ for aerosol part of the profile in Fig. 2a.

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2.2 Aerosol identification algorithm

In order to study the seasonal pattern of the δ of aerosol~~δ~~_{~~aerosol~~}, it is essential to distinguish aerosol from clouds and precipitation. For sites with a co-located cloud radar, one option would be to utilize the Cloudnet classification (Illingworth et al., 2007; Tukiainen et al., 2020), but since not all sites have full Cloudnet instrumentation (cloud radar, ceilometer, microwave radiometer), an algorithm using only Doppler lidar is required.

Using a β' threshold alone is not sufficient for separating aerosol and precipitation. Likewise, using a simple threshold on w alone is not sufficient for differentiating aerosol from light snowfall or precipitation from strong downdrafts. Hence, an algorithm utilizing SNR_{co}, β' and w in both the time and height domain was developed for distinguishing aerosol from larger hydrometeors.

The Aerosol Identification algorithm developed here utilizes 2D-kernel manipulation, which is a commonly used approach in image processing (e.g., Guo et al., 2022; Li et al., 2013; Perreault and Hébert, 2007), to extract various features from the data and to determine the correct class for each data point. A kernel, also referred to as a filter, template, window or mask (Gonzalez and Woods, 2007) is a small 2-D data array. Mathematical operations, such as median, maximum, Gaussian etc., on all values inside the kernel determine its center value. The kernel is run through each data point one by one, replacing its center value with mathematical operations of the neighboring values. ~~The aerosol identification (AI) algorithm developed here utilizes 2D-kernel manipulation to extract various features from the data and to determine the correct class for each data point. The resulting classes are background signal, aerosol, hydrometeor, and undefined. For this algorithm, hydrometeors are defined as cloud (liquid or ice) or precipitation (rain or snow) and do not include aerosol.~~

~~The detailed steps of the algorithm are shown in Figure S3 and explained below:~~

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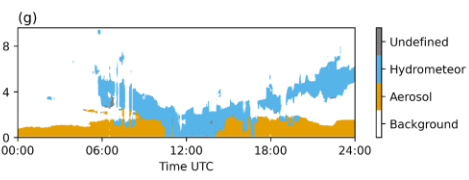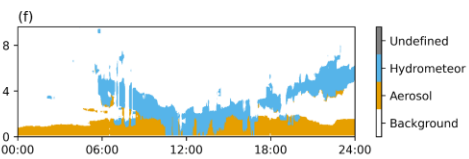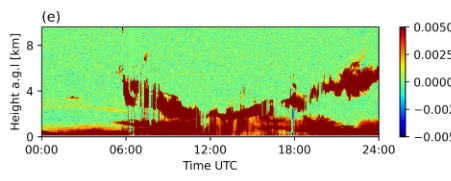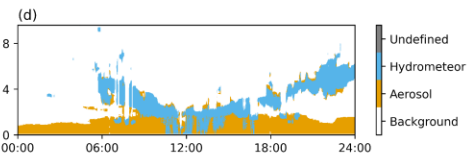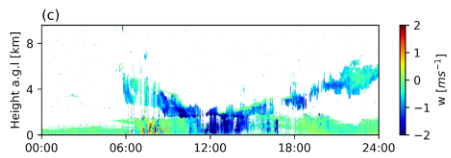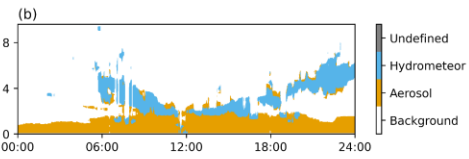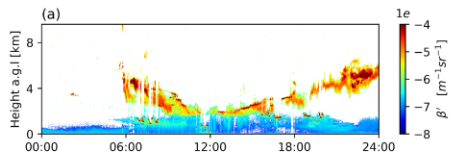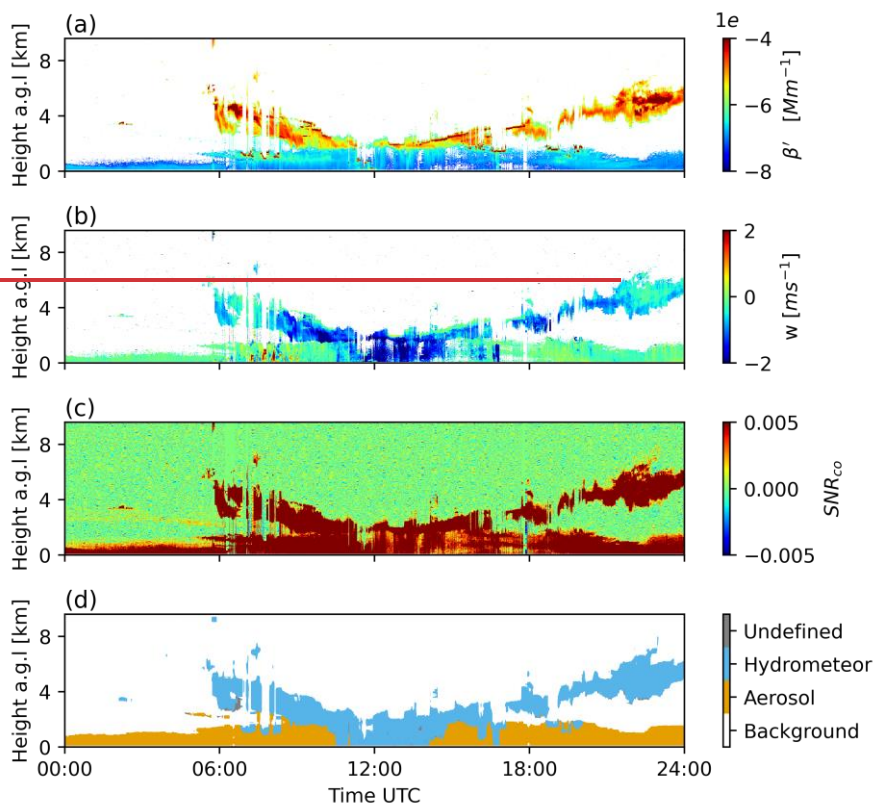
Figure 3: **Atmospheric profiles on 12th of August 2018 at Hyytiälä. The left column displays the measured data where the background noise has been filtered for visualization in [a, c]. a) Attenuated backscatter (β'), c) Vertical velocity (w), e) Signal-to-noise ratio in the co-polar channel (SNR_co). The right column displays the steps from the Aerosol Identification algorithm. b) First step, d) Second step, f) Third step, g) Final step, i.e., the final result.** ~~Data on 12th of August 2018 at Hyytiälä. a) Attenuated backscatter (β'), b) Vertical velocity (w), c) Signal-to-noise ratio in the co-polar channel (SNR_co), d) result from AI algorithm. Data points with SNR_co less than 3 times the background standard deviation of SNR_co has been filtered out for [a, b, c].~~

Figure 3a, 3c, 3e display the measured data from the Doppler lidar in 12th of August 2018 at Hyytiälä. The detail of the Aerosol Identification Algorithm is described in Sect. S3, a brief overview is explained as follows:

1. The first step of the algorithm involves detecting potential hydrometeors and aerosols from background signals based on β' and SNR_co. The result of this step is shown in Fig. 3b.~~Result from this step is shown in Figure S3a~~

~~All data points with SNR_co larger than one standard deviation of the background SNR_co and with β' values less than $10^{-5.5}$ Mm^{-1} are marked as aerosol. A 2D median kernel was then convolved with these aerosol data points to remove noisy signals due to instrumentation and attenuation.~~

~~1. All data points with SNR_co larger than three standard deviations of the background SNR_co and with β' > $10^{-5.5}$ Mm^{-1} were marked as hydrometeor. A 2D median and maximum kernel were then convolved with these hydrometeor data points to remove noisy signals due to instrumentation and attenuation.~~

2. The falling hydrometeor detection step involves separating aerosol in downdrafts due to boundary layer mixing from precipitation using both β' and w. Regions containing both up- and down- drafts are considered to be characteristic of boundary layer mixing, while a regions of continuous downdrafts indicates precipitation. The result from this step is shown in Fig. 3d~~Figure S3b~~.

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- The updraft proxies are identified by selecting data points that have w > 1 m s⁻¹. 2D median and maximum kernels were then convolved to remove noisy signal and expand the updraft proxies.

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- Next, precipitation proxies are selected, having β' > 10⁻⁷ Mm⁻¹ and w < -1 m s⁻¹. A 2D median kernel was then convolved with these data points to remove noisy signals. The precipitation proxies that are in the updraft proxies are then removed.

- All precipitation regions are identified having β' > 10⁻⁷ Mm⁻¹ and w < -0.5 m s⁻¹. A 2D median kernel was then convolved with these data points to remove noisy signals.

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- The updraft region is identified having w > 0.2 m s⁻¹. A 2D maximum kernel was then convolved with these data points to increase the size of the updraft region.

2. Finally, precipitation data points are identified by including the precipitation proxies that overlap with any all precipitation regions but not with updraft regions. A 2D maximum kernel was used for this iterative process.

3. The~~An~~ attenuation correction step ~~step flagged~~sets all observations above clouds and precipitation with their corresponding class since the signal has been heavily attenuated. The result from this step is shown in Fig. 3f.~~Figure S3c~~

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4. ~~In the final step,~~ a fine-tuned aerosol identification process is utilized to improve the aerosol class determination accuracy. First, aerosol clusters are identified using both time and height domain. Then based on the average speed of the aerosol cluster and its connectiveness to the first lidar range gate, it can be classified as either aerosol, hydrometeor or undefined. The final result is shown in Fig. 3g.~~The final result is shown in Figure 3d~~

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4.

5. The resulting classes are background signal, aerosol, hydrometeor, and undefined. For this algorithm, hydrometeors are defined as cloud (liquid or ice) or precipitation (rain or snow) and do not include aerosol. ~~First, aerosol clusters are identified in the time and height domain using the Density-Based Spatial Clustering of Applications with Noise (DBSCAN) algorithm (Ester et al., 1996).~~

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~~6. A cluster is flagged as hydrometeor if the mean w of the cluster < -0.5 m s⁻¹.~~

~~7. The rest of the clusters which are connected to the ground are classified as aerosol.~~

~~8. The rest of the clusters that have w > -0.2 m s⁻¹ are classified as aerosol.~~

~~9. All clusters that do not satisfy of the previous criteria are classified as undefined.~~

270 **2.3 Post-processing**

Aerosol is expected to be well-mixed within each aerosol layer, so in order to extract weak aerosol signal and minimize the random noise, SNR$_{co}$ and SNR$_{cross}$ were averaged for 1 hour ~~before calculating δ$_{aerosol}$~~.

As mentioned before, the SNR data in this study have been processed with the background correction algorithm described by Vakkari et al., (2019). Briefly, the noise floor consists of a non-polynomial component, which is obtained from the background

275 checks according to Vakkari et al. (2019) and a polynomial component, which is obtained from a fit to the aerosol- and hydrometeor- free (background) range gates of each SNR$_{co}$ and SNR$_{cross}$ profile (Manninen et al., 2016; Vakkari et al., 2019).

Typically, the linear part of the noise floor is much larger than the 2nd order polynomial component, but for extended averaging (more than 1 hour) it is essential to include in the background correction. An example of this is shown in Fig. 4a demonstrating how this 2nd order polynomial component can greatly affect the δ of aerosol retrieval in aerosol layers with low SNR (Fig.

280 4d).~~However, at times a weak 2nd order polynomial shape (c.f. Manninen et al., 2016) appears in SNR$_{co}$ and SNR$_{cross}$, when profiles with extended averaging (more than 1 hour) are investigated (Figure 4a). This could greatly affect the δ$_{aerosol}$ retrieval in aerosol layers with low SNR.~~ Previously (Vakkari et al., 2021; Bohlmann et al., 2021), the 2nd order component of the noise floor has been fitted to aerosol- and hydrometeor- free range gates of the SNR profiles based on visual inspection of individual profiles~~this component of the noise floor has been accounted for through a fit to SNR profiles, where hydrometeor and aerosol~~

285 signals have been filtered out through visual inspection. However, given the large number of profiles analysed in this study, this approach is not feasible and thus we have automated the fitting of the 2nd order polynomial, as described below. The fitting algorithm is described in detail in the Sect. S2, and the resulting SNR_{co} and SNR_{cross} profiles are shown in Fig. 4b, 4c. The attenuated backscatter is calculated from the background-corrected SNR_{co}. Next, aerosol layer(s) are identified using the Aerosol Identification algorithmFor a successful fit, aerosol- and hydrometeor- free (background or noise-only) range gates

290 need to be identified in the profile. Firstly, the data is averaged hourly, and the profile SNR is then decomposed by stationary wavelet transform with wavelet bior2.6. Next, the variance of the noise in the SNR is removed by applying a hard threshold shrinkage function using universal thresholding (Donoho and Johnstone, 1994) to the approximation and detail coefficients from level 1 to 4. The SNR is then reconstructed using inverse stationary wavelet transform. Finally, the background range gates are identified as those having reconstructed SNR_{co} values less than the standard deviation of the instrument noise floor

295 divided by the squared root of the number of profiles averaged in that hour. The 2nd order polynomial fit can then be performed on those noise-only data points identified in the profile (Equation 2) and the correction applied to the entire SNR profile, similar to Vakkari et al., (2019).

$$SNR_r = a + h_r \cdot b + c \cdot h_r^2 , \qquad (2)$$

where SNR_r and h_r are the background SNR and height at each range gate, r, and a, b, c are the parameters of the fit for each

300 profile. Overall, the effect of background correction is substantial only for low SNR values as demonstrated in section S2. Next, the aerosol range gates are identified based on the result of the AI algorithm applied to the original non-averaged data. Finally, following Vakkari et al., (2021), the bleed-through corrected δ in aerosol regions $\delta_{aerosol}$ is calculated as

$$\delta \delta_{aerosol} = \frac{SNR_{cross} - B \cdot SNR_{co}}{SNR_{co}} , \qquad (33)$$

305 where B is the estimated bleed through of each instrument (Table 3)obtained from section 2.1.2.

The resulting δ is shown in Fig. 4d, and the estimation of its uncertainty is presented in Sect. S2. Additionally, the post-processed δ of aerosol was collected from the whole dataset and compared with the original δ. The result described in Sect. S2 shows that the post-processing procedure substantially improved the δ of aerosol with low SNR valuesThe uncertainty of the resulting $\delta_{aerosol}$ is presented in section 2.3.1.
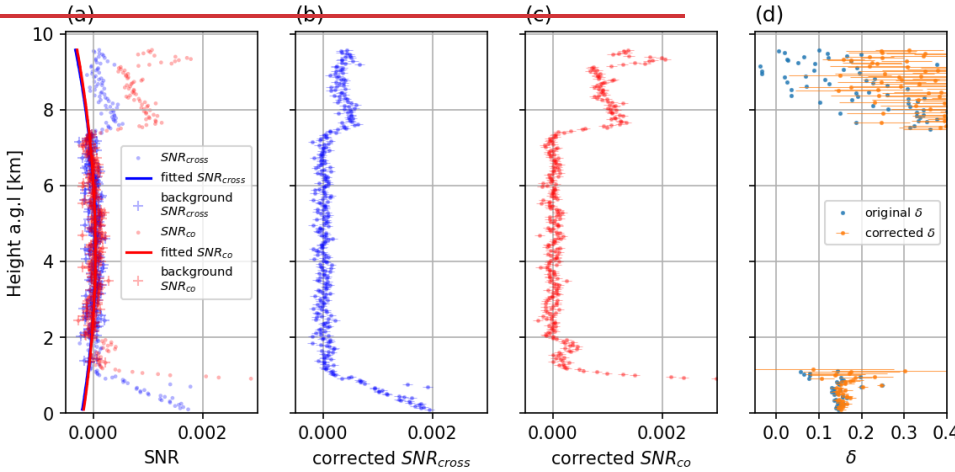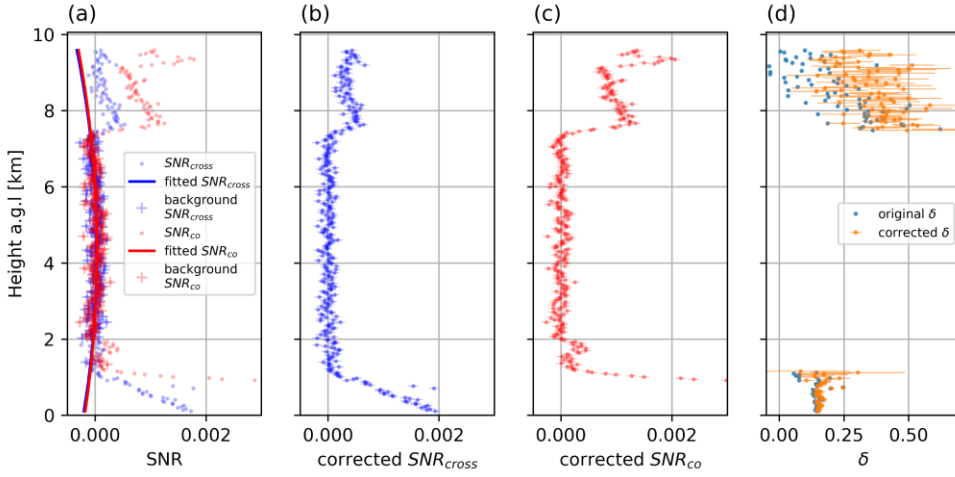
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Figure 4: An example results of the post-processing procedure ~~Results of background correction~~ on 2019-08-30 at 13:00 to 14:00 UTC at Hyytiälä with instrument 46. a) Profiles of co- and cross-polar signal-to-noise ratio (SNR_{co} and SNR_{cross})~~,~~ where aerosol- and hydrometeor-free regions and the corresponding 2nd order polynomial fits are indicated. b) Corrected SNR_{cross} profile with error bar representing the fitting uncertainty. c) Corrected SNR_{co} profile with error bar representing the fitting uncertainty. d) δ profile before and after the 2nd order fit correction. ~~The uncertainty in $\delta_{aerosol}$ is described in section 2.3.1.~~

~~**2.3.1 Uncertainty in $\delta_{aerosol}$**~~

~~The background and the bleed through corrections include uncertainties that will propagate through to the uncertainty in the final retrieved $\delta_{aerosol}$. A Bayesian approach utilizing Stan (Carpenter et al., 2017) has been used to estimate these uncertainties. The full Bayesian inference is obtained through a sampling method based on Hamiltonian Monte Carlo simulation (Betancourt and Girolami, 2015), which is a form of Markov chain Monte Carlo sampling. The resulting posterior distribution is used to estimate the mean and uncertainty of the parameters and generate the posterior predictive distribution of the variables.~~

~~The 2nd order polynomial model of SNR is similar to equation (2), but with the error component added:~~

$$SNR_x = a + h_x \cdot b + c \cdot h_x^2 + \epsilon,\ \ \ \ \ \ \ \ \ \ (4)$$

~~where $\epsilon \sim Normal(0, \sigma^2)$.~~

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~~Equation (4) is equivalent to $SNR_x \sim Normal(a + h_x \cdot b + c \cdot h_x^2, \sigma)$, or the so-called likelihood $p(SNR \mid a,b,c,\sigma,h_x)$. The standard non-informative prior was used for all parameters:~~

$$p(a) \sim Normal(0, 10^3)$$
$$p(b) \sim Normal(0, 10^3)$$
$$p(c) \sim Normal(0, 10^3)$$
$$p(\sigma) \sim Gamma^{-1}(0.001, 0.001)$$

~~The posterior distributions of the parameters are then obtained by sampling through Stan.~~

$$p(a,b,c,\sigma \mid SNR, h) \propto p(SNR \mid a,b,c,\sigma) \, p(a,b,c,\sigma)$$

~~Four different sampling chains are utilized with 2000 samples for each chain (excluding the 500 warmup samples), and convergence diagnostics have been carried out to ensure that the chains converged. The sampling results represent the posterior distribution of all the parameters.~~

~~From these posterior distributions, the mean and standard deviation of the parameters can be calculated. The fitted background SNR distribution at all heights can be generated by sampling from the posterior predictive distribution. It is done through selecting random draws from the posterior, which are then plugged into equation (4) to calculate the posterior predictive samples. The mean and standard deviation of the fitted background SNR at each height can be estimated by calculating the mean and standard deviation of the generated samples at each height.~~

~~The samples of the corrected SNR distribution at each height are calculated by dividing the original SNR by the samples from the estimated background SNR distribution at each height. The samples of the final δ_aerosol distribution at each height are then calculated following equation 2 assuming the bleed-through follows a normal distribution with mean and standard deviation estimated from section 3.1.2. The mean and uncertainty of the final δ_aerosol is estimated by calculating the mean and standard deviation of the samples at each height.~~

2.4 Air mass origin

For two case studies, the air mass origin was investigated using the Lagrangian transport model Flexpart version 10.4 run in backwards mode (Seibert and Frank, 2004; Stohl et al., 2005; Pisso et al., 2019). Flexpart was run using ERA5 reanalysis obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) as the meteorological input. ERA5 was obtained with a temporal resolution of 1 hour and a spatial resolution of 0.25° for a domain covering 125° W to 75° E and 10° N to 85° N. Vertical model levels 50-137 were retrieved, which includes approximately the lowest 20 km a.g.l. For the elevated layers considered in the case studies, Flexpart was run backwards in time for 7 days and potential emission sensitivity (PES) was saved with 1-hour temporal, 0.2° latitude-longitude resolution. The output resolution in the vertical was 250 m from ground level to 5 km a.g.l. Above 5 km a.g.l, output levels at 10 km a.g.l and 50 km a.g.l were included. PES is proportional to the air mass residence time in a grid cell and was obtained in units of seconds (Seibert and Frank, 2004; Pisso et al., 2019).

3 Results and discussion

3.1 Instrumental performance

3.1.1 Noise floor level

Figure 5a displays the noise floor level for each instrument, calculated as the standard deviation of the SNR_{co} (σ_{SNR}) in the aerosol- and hydrometeor- free region. As expected, integration time is one important factor with, for example, instrument Utö-32 showing a dramatic increase in the noise floor level in summer 2016 when the integration time was decreased (from 27.5s to 5s). Since each instrument may have its own configuration, normalization of the noise floor level considering such differences is required in order to compare the noise floor between instruments.

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Figure 5b illustrates the noise floor levels calculated after scaling for number of pulses in each integration time and demonstrates that most instruments remained stable during the study period. Fluctuations were observed in the noise floor level for the old instrument Utö-32, but the noise floor level itself is deemed to be low, with visual inspection indicating no issues in the data quality. Sodankylä-54 is the only instrument that shows a systematic increase in the noise floor level over time. This is a worrying sign, but so far, the increase in the noise floor has been relatively small at approximately 2% from 2017 to 2019. However, further increases in the noise floor will begin to limit the retrieval of weak signals.

The StreamLine Pro lidar instruments, Vehmasmäki-53 and Sodankylä-54 have similar and systematically higher noise floor levels than the other instruments. This is due to the Streamline Pro models were configured to utilize only half of the bandwidth, i.e half the Nyquist velocity. Assuming the noise is thermal noise and evenly distributed across the frequency spectrum, reducing the bandwidth by half will reduce the noise power by half (Nyquist, 1928; Johnson, 1928). The Utö-32XR instrument, which is the Utö-32 system after being upgraded with a StreamLine XR transmitter and receiver, has the lowest noise floor of all instruments in this study.
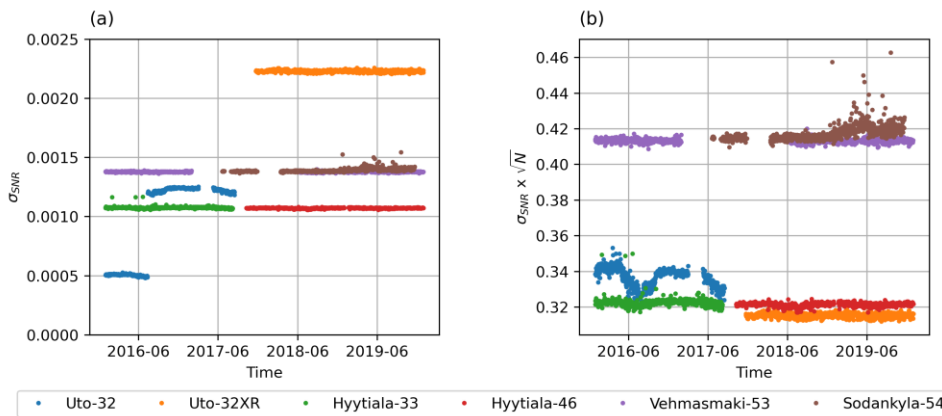

Figure 5: (a) Standard deviation of the background co-polar signal-to-noise ratio (σ_{SNR}). (b) The same but scaled with number of pulses in each integration (denoted with N).

3.1.2 Bleed through

Figure 6 displays the time series and the distribution of δ at liquid cloud base for each instrument in the network. The time series is used to assess the stability of the internal polarizer over time and the distribution is used to calculate the bleed-through (B) and its uncertainty. Overall, there is no significant trend in the bleed-through of all the instruments.

For Utö-32 (Figure Fig. 6a), the overall mean bleed-through δ is the highest of all the instruments in this study. This probably stems from its long integration time configuration (see Table 2), resulting in SNR_{co} and SNR_{cross} not measuring the same part of the cloud.

In addition, the occurrence of liquid cloud varies in time and with location. Mixed-phased clouds are difficult to distinguish from liquid-only clouds, hence, the high value of δ at liquid cloud base especially during wintertime (Figure Fig. 6) might be due to the misclassification with mixed-phased clouds. This effect is more prominent in Utö-32 and Hyytiälä, leading to longer tailed distributions (Fig. Figure 6c and 6d) than at other sites. On the other hand, for Utö-32XR, Vehmasmäki-53 and Sodankylä-54 (Fig. Figure 6b, 6e and 6f), the δ at the collected liquid cloud base varies much less and remain stable at a low level (δ <0.01) throughout the year.

To quantify the bleed-through of the internal polarizer, the elevated value of δ due to imperfect sampling of pure liquid clouds need to be excluded. Here, we assume that for Utö-32, Hyytiälä-33 and Hyytiälä-46, the tail of the distribution due to mixed-

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phase clouds or non-ideal sampling of liquid clouds, whilst the peak of the distribution is from the liquid cloud base. A Gaussian mixture model, which is a variant of finite mixture models (Baek et al., 2010), was used to derive the mean and standard

400 deviation of δ at the liquid cloud base ignoring the tail of the distribution. For most instruments, excluding the tail of the distribution in Figure. 6 has a minimal effect on the average bleed-through estimate, which remains around 1 % as shown in Table 3. The largest effect is seen for Hyytiälä-46, where including all δ values at liquid cloud base results in a bleed-through estimate 0.020 ± 0.022, while excluding the tail decreases the bleed-through estimate to 0.008 ± 0.007. Overall, the values of the best-estimate bleed-through in this study are smaller than the previous short case study estimates of 0.011 ± 0.007 in

405 Limassol (Vakkari et al., 2021); 0.016 ± 0.009 and 0.013 ± 0.006 in Vehmasmäki (Vakkari et al., 2021; Bohlmann et al., 2021) for similar instruments.
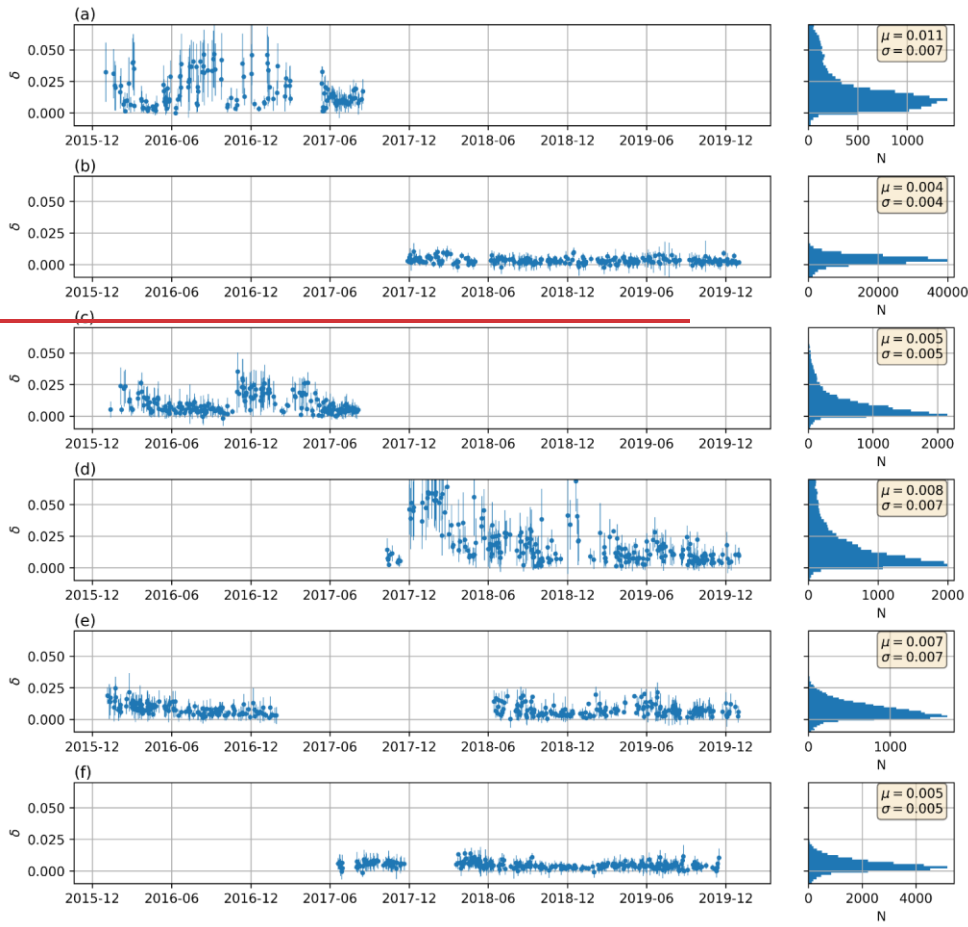
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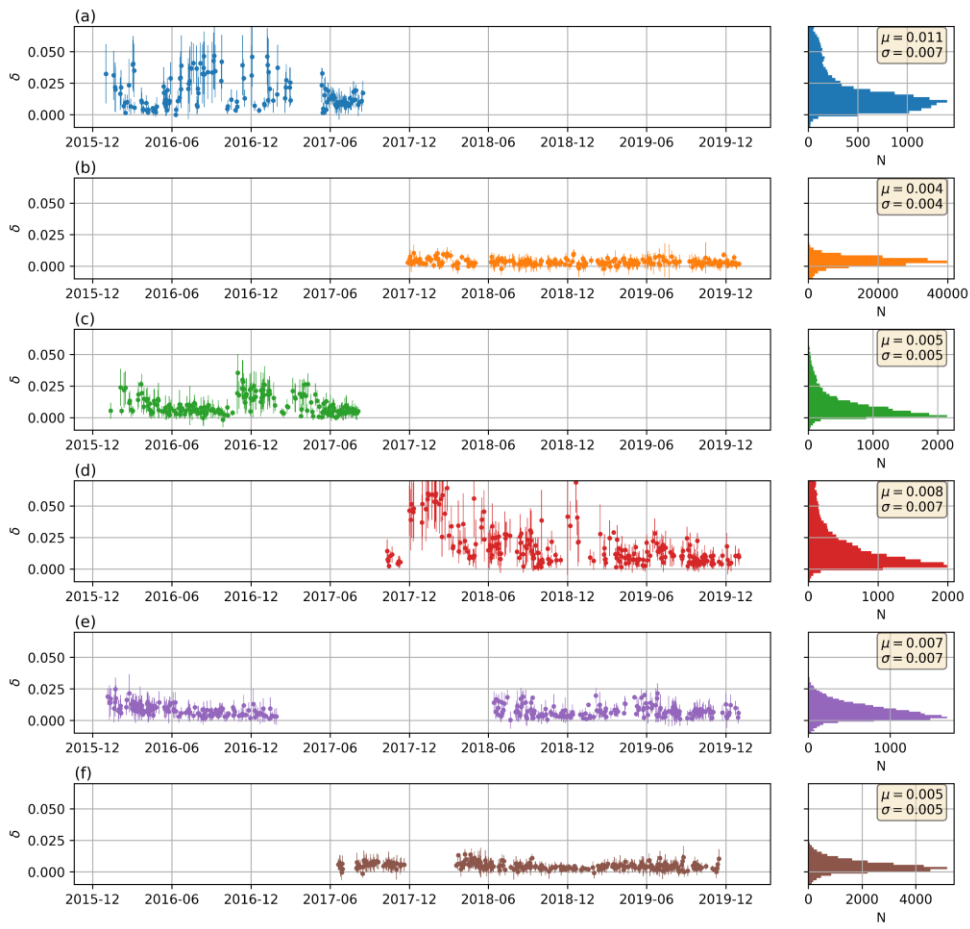
Figure 6: Time series (left panels) and histograms (right panels) of depolarization ratio (δ) at liquid cloud base in a) Utö-32, b) Utö-32XR, c) Hyytiälä-33, d) Hyytiälä-46, e) Vehmasmäki-53, f) Sodankylä-54. The best estimates of mean (μ) and standard deviation (σμ) of δ at the liquid cloud base in each site are shown in the right panel.

Instrument	Best estimate of bleed-through	Bleed-through including all data
Utö-32	0.011 ± 0.007	0.018 ± 0.018
Utö-32XR	0.004 ± 0.004	0.004 ± 0.004
Hyytiälä-33	0.005 ± 0.005	0.009 ± 0.011
Hyytiälä-46	0.008 ± 0.007	0.020 ± 0.022
Vehmasmäki-53	0.007 ± 0.007	0.007 ± 0.007
Sodankylä-54	0.005 ± 0.005	0.004 ± 0.005

Table 3: Bleed-through (mean and standard deviation of depolarization ratio at the liquid cloud base) for each instrument.

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3.2 Comparing Aerosol Identification algorithm with Cloudnet classification

Cloudnet's goal is to provide continuous ground-based observations of the cloud properties for forecast and climate model (Illingworth et al., 2007). On the other hand, the primary aim of the Aerosol Identification (AI) algorithm developed here is to exclude hydrometeors from the aerosol mask to ensure that aerosol characteristics are not biased by signal from hydrometeors.

Of all the sites in this study, only Hyytiälä belongs to the Cloudnet's network. There, the result of the Aerosol Identification~~AI~~ algorithm based on Doppler lidar only (~~AI algorithm,~~ Sect. 2.2) can be compared with the classification from Cloudnet (Illingworth et al., 2007; Tukiainen et al., 2020). However, there are marked differences between the two algorithms. The Cloudnet classification algorithm utilises a combination of several instrument, most importantly being lidar and cloud radar (Illingworth et al., 2007). The backscattered signal from radars is much more sensitive to the diameter (D) of the particle, $\sim D^6$ (Rauber and Nesbitt, 2016) compared to lidar with only $\sim D^2$ (Weitkamp, 2005). As the result, radar's signal is dominated by larger particles while lidar's is dominated by smaller particles with higher concentration. Additionally, the extinction of radar signal in non-precipitating clouds is negligible, so it can observe much further into the clouds.

Figure 7 presents a side-by-side comparison example of the results from these algorithms. At first glance, it is obvious that Cloudnet underestimates the extent of the aerosol layer compared to the Aerosol Identification ~~AI~~ algorithm. This is due to the improved post-processing procedure applied to the Doppler lidar data, which allows the Aerosol Identification ~~AI~~ algorithm to capture data from weaker aerosol signal than Cloudnet. Unsurprisingly, cloud mask in Cloudnet classification includes the full cloud layer, while Aerosol Identification ~~AI~~ algorithm indicates often only the cloud base due to the attenuation of lidar signal. There are some minor differences in the precipitation zone such as right before 18:00 UTC in ~~Figure~~ Fig. 7a and 7b and at 18:00 UTC in Fig. ~~Figure~~ 7e and 7f, when some parts of the precipitation in Cloudnet are flagged as aerosol in the Aerosol Identification~~AI~~ algorithm. This is probably due to the differences in sensitivity of different particle size in cloud radar and lidar as discussed previously. Figure 7g and 7h display a snow event from 0:00 UTC to around 11:00 UTC. Cloudnet shows that some parts of snowfall eventually melt into raindrops near the ground, whilst the Aerosol Identification ~~AI~~ algorithm ~~also~~ correctly classified the whole event as hydrometeor.

After comparing all the data in Hyytiälä throughout the whole study period, we found that 7.7% of the aerosol data points from the Aerosol Identification ~~AI~~ algorithm is classified as hydrometeor in Cloudnet algorithm. This result shows that the Aerosol Identification ~~AI~~ algorithm performs adequately in extracting aerosol data comparing to Cloudnet classification algorithm. The differences between these two algorithms stem from differences in goals and instrumentations. Given the large amount of data, we are confident that the Aerosol Identification ~~AI~~ algorithm is capable of extracting the overall statistics of the δ of aerosol ~~$\delta_{aerosol}$~~ for the purpose of this study.
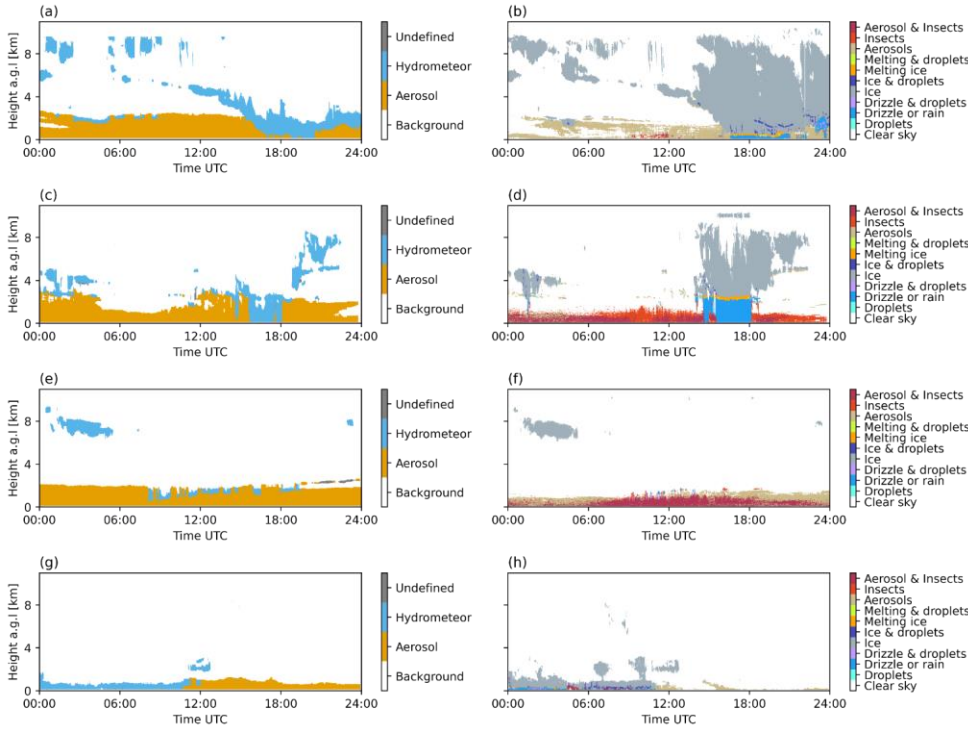
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Figure 7: Results in Hyytiälä from the aerosol identification algorithm and Cloudnet classification algorithm respectively on [a, b] 2018-06-11; [c, d] 2019-04-07; [e, f] 2019-08-07; and [g, h] 2019-12-31.

3.4 3 Aerosol particle depolarization ratio

3.3.1 Case studies

Here, we present two case studies of elevated aerosol layer observed by the lidars in Southern Finland. The δ of aerosol was obtained following the post-processing procedure described in Sect. 2.3

Hyytiälä and Utö in April 2018

From late night 14th of April 2018 to morning 15th of April 2018, an elevated aerosol layer between 2 km to 4 km a.g.l. was observed above Hyytiälä (Fig. 8a, 8b). The average profiles (Fig. 8e, 8f) from 03:00 to 04:00 UTC on 15th of April 2018 show an elevated aerosol layer with the δ of aerosol at 0.24 ± 0.008, while in the boundary layer (from the surface to 1.5 km a.g.l.), the δ of aerosol is at 0.12 ± 0.004. Similarly, an elevated aerosol layer between 2 km to 3 km was also observed in Utö (Fig. 8c, 8d). The averaged profiles (Fig. 8e, 8f) from 00:00 to 01:00 UTC on 15th of April show that the δ of this layer is at 0.226 ± 0.005 while in the boundary layer (from the surface up to 2km), the δ of aerosol is between 0.05 and 0.1. These substantial difference in the δ of aerosol between the elevated layer and the boundary layer indicates that the elevated aerosol had likely undergone long-range transport to these sites.

The FLEXPART simulation of this layer (Fig. S5.3) indicates that the air mass was not in contact with the surface for 4.5 days prior to arrival over Hyytiälä. However, 5-7 days before arrival, a contribution from the surface layer was seen coming mostly from the western Sahara, with minor contributions from the Mediterranean. On 15th April an overpass by CALIPSO crossed

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the air mass transport pathway relatively close to the observation sites in Finland at 62.8N, 24.3E and the CALIOP aerosol subtype V4.2 product indicates a dust layer at 3-5 km above ground at this location (Fig. S5). On 15 April also dust aerosol optical depth at 550nm from the CAMS model (Benedetti et al., 2009; Morcrette et al., 2009) indicates presence of dust over south western Finland (Fig. S6).

These results suggest the layer to be most likely Saharan dust, although the observed δ is slightly lower than the value of 0.30 reported by Vakkari et al., (2021) for dust at 1565 nm. This could be due to gravitational settling of large dust particles during long range transport (Haarig et al., 2017b).
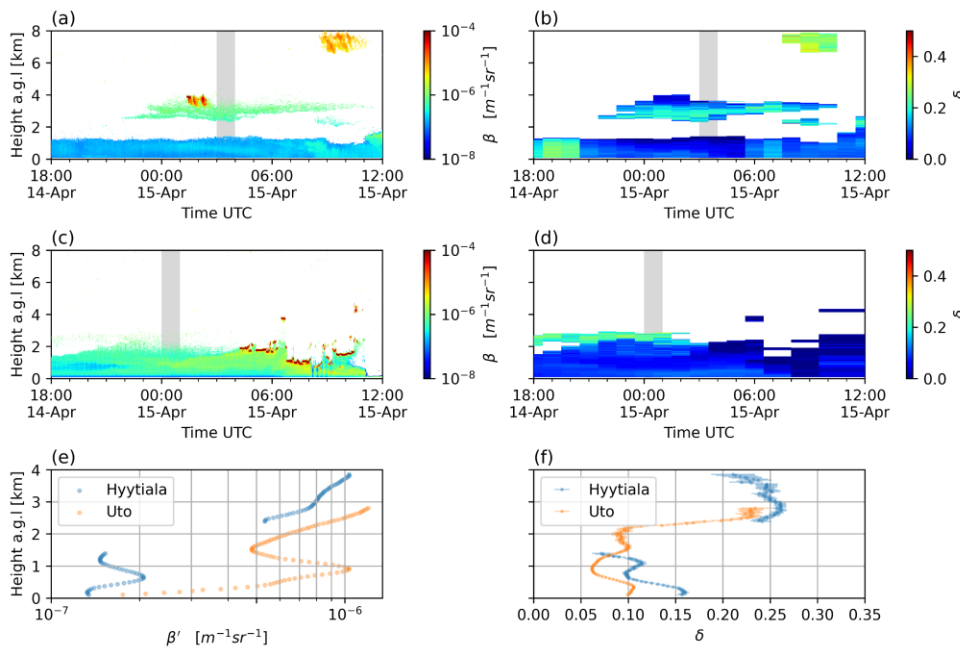
Figure 8: Profiles from 2018-04-14 to 2018-04-15 in Hyytiälä and Utö. a) Attenuated backscatter (β') in Hyytiälä, b) 1-hour averaged depolarization ratio (δ) in Hyytiälä. c) Attenuated backscatter (β') in Utö, d) 1-hour averaged depolarization ratio (δ) in Utö. And 300m running mean (in shaded area) on 2018-04-15 03:00-04:00 UTC in Hyytiälä and 2018-04-15 00:00-01:00 UTC in Utö of e) β' profiles f) δ profiles.

Utö in May 2017

On 13th of May 2017, an elevated aerosol layer between 800 m a.g.l and 2 km a.g.l was observed over Utö (Fig. 9). For the period on 2017-05-13 from 18:00 to 20:00 UTC, the δ of this elevated aerosol layer is at 0.23 ± 0.01 and decreases with height (Fig. 9d and 9f). At the same time, the δ of the boundary layer aerosol from the surface up to 500 m is at 0.26 ± 0.009.

The air mass history shows that this layer originated from both offshore in the Baltic Sea and from continental Finland (Fig. S6). The layer was elevated from the surface only 1 day before arrival and remained at a height of 1-2 km a.g.l. In this region there are no other known sources of aerosol with such high δ except for pollen, and thus this layer is probably pollen transported from the surrounding continental areas. From 2017-05-14 00:00-05:00 UTC, this layer merged into the boundary layer, with the maximum value of δ in the boundary aerosol layer increasing to 0.30 ± 0.007. This value of δ is comparable to previous observations at the same wavelength for spruce pollen in boreal forest; 0.269 ± 0.005 (Vakkari et al., 2021) and 0.29 ± 0.10 (Bohlmann et al., 2021).
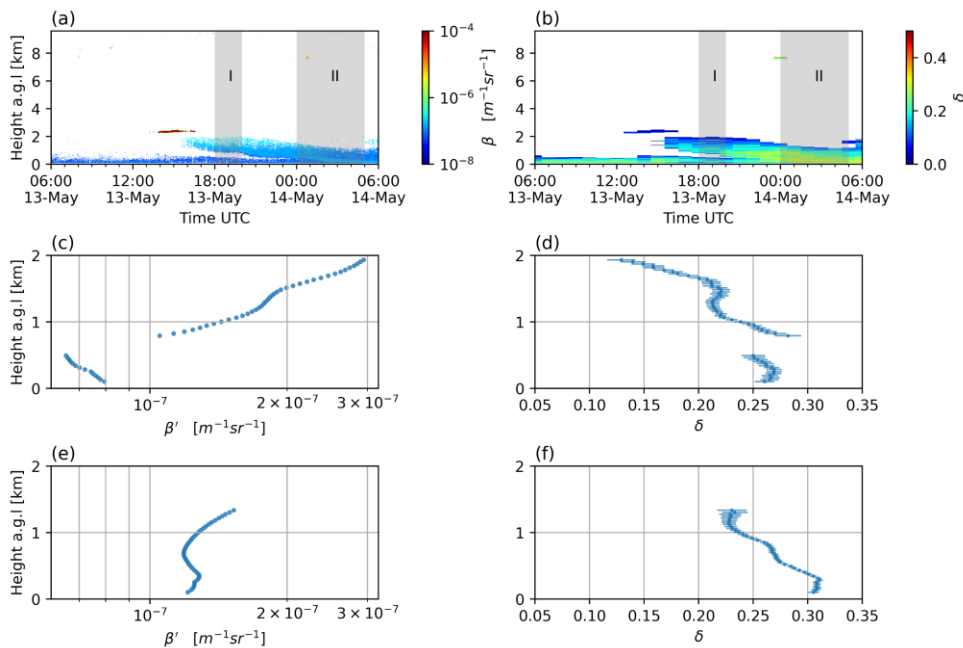
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Figure 9: Profiles on 2017-05-13 to 2017-05-14 at Utö a) Attenuated backscatter (β') b) 1-hour averaged depolarization ratio (δ). And 300m running mean (shaded area I) at 2017-05-13 18:00-20:00 UTC of c) β' profile c) δ profile. 300 m running mean (shaded area II) at 2017-05-14 00:00-05:00 UTC of c) β' profile c) δ profile.

3.3.2 Long-term analysis

495   Here, we analyse the δ in aerosol region (δ of aerosol) $\delta_{aerosol}$ from the four-year observational data set at four sites in Finland. The δ of aerosol $\delta_{aerosol}$ was obtained following the post-processing procedure described in Sect. 2.3 Section 2.3 and the Aerosol Identification AI algorithm (Sect. 2.2) was used to exclude hydrometeors from the data set. All data that have standard deviation of the δ of aerosol $\delta_{aerosol}$ larger than 0.05 have been filtered out.

Sites	25th percentile	50th percentile	75th percentile	Mean	Standard deviation
Utö	0.03	0.07	0.12	0.09	0.07
Hyytiälä	0.06	0.11	0.18	0.13	0.08
Vehmasmäki	0.05	0.10	0.16	0.11	0.07
Sodankylä	0.01	0.05	0.11	0.07	0.08

500   **Table 4: Overall statistics of δaerosol across all the sites**

Overall, as seen in Table 5, the average δ of aerosol $\delta_{aerosol}$ isare higher in the boreal forest sites of Hyytiälä (0.13 ± 0.08) and Vehmasmäki (0.11 ± 0.07) compared to Utö (0.09 ± 0.07) and Sodankylä (0.07 ± 0.08). This can be explained by the abundance of pollen with high δ$\delta_{aerosol}$ in the boreal forest sites (Aaltonen et al., 2012; Shang et al., 2020; Vakkari et al., 2021; Bohlmann et al., 2021). Located in the marine site, the lidar at Utö observes a high fraction of marine aerosols with low δ, $\delta_{aerosol}$ as also
505   found in previous studies (Groß et al., 2011, 2013; Haarig et al., 2017a; Vakkari et al., 2021; Mylonaki et al., 2021). In Sodankylä, which is a clean rural subarctic environment, the δ of aerosol $\delta_{aerosol}$ is found to be the lowest of the sites considered here.
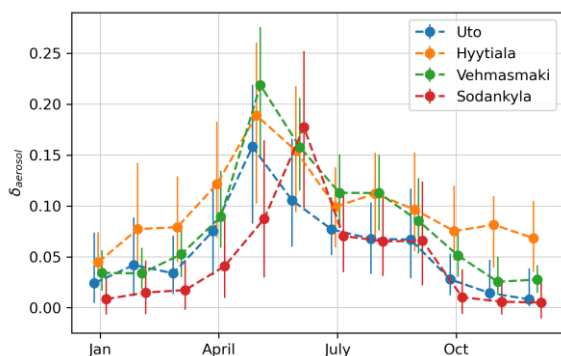
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Figure 810: Monthly median of the particle aerosol depolarization ratio of aerosol (δaerosol) across all the sites, error bars show the 25th and 75th quantile

Figure 8 10 shows the average monthly median of the δ of aerosol δaerosol for all sites. Overall, the δ of aerosol δaerosol is increased in the summer and remains low in the winter. The enhanced values of the δ of aerosol δaerosol from April to the end of September coincide with the growing season of the boreal forest (Manninen et al., 2014) when airborne pollen and other particles of biological origin are abundant in Finland. The most common sources of pollen in the Finnish boreal forest are birch (Betula), spruce (Picea), pine (Pinus), and nettle (Urtica). They contribute to more than 90% of the total pollen load (Shang et al., 2020; Manninen et al., 2014). The peaks of the δ of aerosol δaerosol occurs in the pollen season of birch, spruce, and pine (Shang et al., 2020) between May and early June. The irregular shape of these pollen types has been observed to have high δ such as 0.10 ± 0.06 for birch at 532 nm (Bohlmann et al., 2019), 0.29 ± 0.10 for spruce at 1565 nm (Bohlmann et al., 2021), and 0.36 ± 0.01 for pine at 532 nm (Shang et al., 2020). The relatively high concentration of these pollen grains in the atmosphere likely explains the peak of the δ of aerosol δaerosol in May for the boreal forest sites of Hyytiälä and Vehmasmäki.

However, the peak in the δ of aerosol δaerosol at Sodankylä occurs one month later than the other sites. Due to its location in the sub-arctic, the snow melt and the beginning of the pollen season start later at around June in Sodankylä than at other sites in central and southern Finland (Koivikko et al., 1986; Oikonen et al., 2005). Interestingly, despite its marine location, the δ of aerosol δaerosol in Utö also peaks in May and the δ of aerosol δaerosol value remains high during summer, probably due to transported pollen from the mainland. Even though pollen grains are typically large from 10 to 100 μm (Manninen et al., 2014), they are low-density particles, which make them prone to be lifted by turbulent air flows up to several kilometres and be dispersed by the wind (Bohlmann et al., 2019). Several studies (Rousseau et al., 2003, 2006; Skjøth et al., 2007; Szczepanek et al., 2017) have demonstrated the long-range transport of pollen over thousands of kilometres. Additionally, marine aerosol at lower relative humidity in the summer (see Figure. 120) could contribute to the higher δ of aerosol δaerosol in the summer at Utö. Haarig et al., (2017a) found that at low relative humidity, elevated marine aerosol can crystalize and become mostly cubic-like in shape (Wise et al., 2007), which results in a higher δ of aerosolδaerosol up to 0.15 ± 0.03 at 532 nm and 0.1 ± 0.01 at 1064 nm.

During the winter months from October to March, the δ of aerosol δaerosol remains low across all sites. Luoma et al., (2019) found that there is a high fraction of aerosol from anthropogenic sources and domestic wood burning in the winter in Hyytiälä, and these are probably major sources of aerosol in other continental sites as well. Burton et al., (2015) reported the δ δaerosol of smoke as 0.019 ± 0.005 at 1064nm, and with its negative wavelength dependency (Haarig et al., 2018; Ohneiser et al., 2020), the δ δaerosol of smoke at 1565nm would probably have even smaller values. Anthropogenic aerosol has also been found to have small δδaerosol values < 0.05 at 532nm (Müller et al., 2007; Burton et al., 2013; Mylonaki et al., 2021). The low δ of aerosolδaerosol in Utö < 0.05 agrees with the typical δ δaerosol of wet and polluted marine aerosol found at various wavelengths (Groß et al., 2011; Haarig et al., 2017a; Vakkari et al., 2021; Mylonaki et al., 2021).
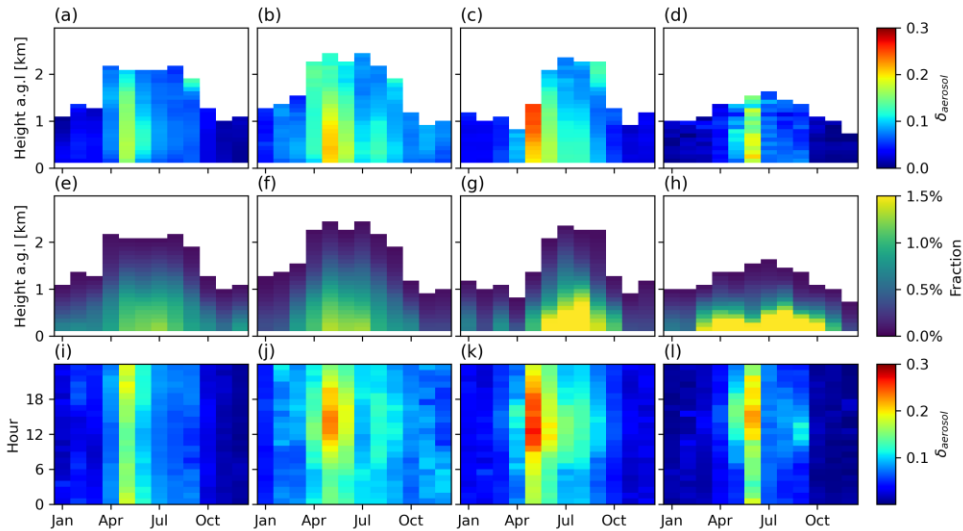
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Figure ~~9~~11: Median of ~~aerosol depolarization ratio~~the particle depolarization ratio of aerosol ($\delta_{aerosol}$) profiles across all the sites. Utö [a, e, i]; Hyytiälä [b, f, j]; Vehmasmäki [c, g, k]; and Sodankylä [d, h, l]. Distribution of $\delta_{aerosol}$ with height ~~ratio~~ in each month is shown on the first row [a, b, c, d]. The frequency of observations relative to height in each month is shown on the second row [e, f, g, h]. The last row [i, j, k, l] displays the diurnal pattern of ~~a~~ $\delta_{aerosol}$ in each month.

Figure ~~9~~11a, ~~9~~11b, ~~9~~11c and ~~9~~11d display the average δ of aerosol ~~$\delta_{aerosol}$~~ as a function of height for each site. During summer from April to August, aerosols with high ~~δ~~$\delta_{aerosol}$ are concentrated within the boundary layer, often below 1 km a.g.l. However, in Vehmasmäki during May, the δ of aerosol ~~$\delta_{aerosol}$~~ increases with height, ~~the $\delta_{aerosol}$~~it ~~value~~ can reach up to 0.3 at 1.2km a.g.l. As mentioned, pollen grains can get easily lofted to higher altitude and since the air is dryer at high altitude, pollen might fold onto itself to prevent further dehydration which would lead to higher δ ~~$\delta_{aerosol}$~~ (Bohlmann et al., 2019).

In September at Utö, Hyytiälä and Vehmasmäki, aerosol with ~~δ~~$\delta_{aerosol}$ > 0.1 can be observed at around 2 km a.g.l. This is probably due to transported aerosol since the δ of aerosol ~~the $\delta_{aerosol}$~~ is much higher than boundary layer aerosol near the ground. During winter months from October to March, the δ of aerosol distributed ~~$\delta_{aerosol}$ values were distributed~~ uniformly at all heights for every site.

Figure ~~9e~~11e, ~~9~~11f, ~~9~~11g and 11~~9~~h show the frequency of aerosol detected at each height level across all the sites. Overall, more aerosol is observed in the summer months, and at higher altitude, due to the higher boundary layer height in summer. In Sodankylä, aerosol is observed less frequently at altitudes above 1 km a.g.l compared to other sites.

Figure ~~9i~~11i, ~~9j~~11j, ~~9k~~11k and ~~9l~~11l illustrate the diurnal pattern of the δ of aerosol ~~$\delta_{aerosol}$~~ across all the sites. During May-June, when the monthly δ of aerosol ~~$\delta_{aerosol}$~~ is at its highest, the median hourly δ of aerosol ~~median $\delta_{aerosol}$~~ in Hyytiälä, Vehmasmäki and Sodankylä peaks in the afternoon. The release of pollen is positively influenced by higher temperature (Bartková-Ščevková, 2003), so the higher δ of aerosol ~~$\delta_{aerosol}$~~ during daytime might be due to higher fraction of irregular shaped pollen being released at noon. Simultaneously, higher mixing layer height during afternoon hours enables more pollen grains released at the surface to be lifted above the lidar's minimum range of 90m (see Table 2). This result also agrees with the previous findings (Noh et al., 2013a; Bohlmann et al., 2021), where the high ~~δ~~$\delta_{aerosol}$ from pollen is only observed in the boundary layer during daytime.

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On the contrary, during May in Utö, the δ of aerosolδaerosol peaks at night and remains low during the day (~~Figure~~ Fig. ~~9i~~11i). We attribute this high δ of aerosol value ~~value of δaerosol~~ to the transported pollen arriving from the continent at night, which may be part of the returning flow of a sea breeze circulation in the Baltic Sea (Dailidė et al., 2022).

3.4.1 The effect of relative humidity

Studies show that relative humidity (RH) can change the shape of marine aerosol (Granados-Muñoz et al., 2015; Haarig et al., 2017a) and pollen grains (Franchi et al., 1984; Katifori et al., 2010; Griffiths et al., 2012) through hygroscopic growth or rupture into smaller fragments (Taylor et al., 2002, 2004; Miguel et al., 2006; Hughes et al., 2020; Bohlmann et al., 2021). Hence, the δ of aerosol ~~δaerosol~~ at the sites in this study may also respond to the ambient RH. However, as RH profiles are not measured, we investigate the connection between the surface RH at 2 m a.g.l and the δ of aerosol ~~δaerosol~~ below 300 m a.g.l. Figure 1~~0~~2 shows the seasonal and diurnal pattern of the δ of aerosolδaerosol and RH below 300 m a.g.l. The δ of aerosol ~~δaerosol~~ pattern of the closest 300m (~~Figure~~ Fig. 1~~0~~2e, 12~~0~~f, 12~~0~~g and 12~~0~~h) is similar to that of the whole profile (~~Figure~~ Fig. 119i, 119j, 119k, 119l) i.e., ~~in that~~ its value is highest in the summer and lowest in winter. During May and June, the continental sites present a strong diurnal variation in both RH and the δ of aerosolδaerosol. However, the island of Utö also shows a similar if less pronounced diurnal cycle in RH to the continental sites, but an opposite diurnal cycle response for the δ of aerosolδaerosol as noted before due to transported pollen.

Previously, pollen concentrations have been shown to be enhanced around noon when RH is at its lowest (Käpylä, 1984; Latorre and Caccavari, 2009; Noh et al., 2013b). In fact, pollen release is highly dependent on the particular ambient weather conditions including rainfall, air temperature, RH, duration of sunshine and wind speed (Gilissen, 1977; Käpylä, 1984; Jato et al., 2000; Alba et al., 2000; Adams-Groom et al., 2002; Bartková-Ščevková, 2003; Vázquez et al., 2003; Noh et al., 2013b). Moreover, low RH could affect the diffusion of pollen in the atmosphere by decreasing the specific gravity of pollen grains, thus negatively reduce their ability to settle to the ground (Durham, 1943), i.e., be more easily diffused above the lidar minimum range of 90 m a.g.l.
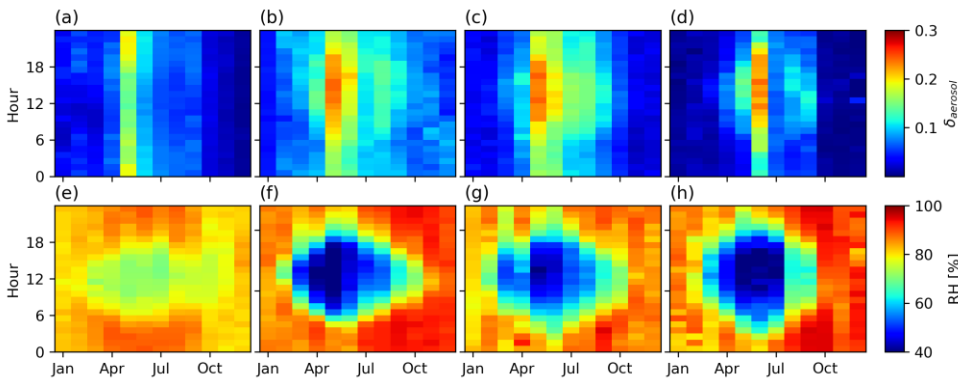
Figure ~~10~~12: Diurnal pattern of aerosol depolarization ratio (δaerosol) from 90 m to 300 m a.g.l and relative humidity (RH) at 2 m a.g.l ~~below 300m a.g.l~~ respectively in Utö [a, e], Hyytiälä [b, f], Vehmasmäki [c, g], Sodankylä [d, h]

Statistical analysis (Table S4) shows an overall negative correlation between RH and the δ of aerosolδaerosol. However, without additional data, attributing the change of aerosol physical properties (such as hygroscopic growth) as the reason for this negative correlation is not possible. The change of aerosol type such as from pollen in the summer to anthropogenic aerosol in the winter, or diurnal changes in pollen release could be the main driver of the δ of aerosolδaerosol.

3.5 Case studies

3.5.1 Hyytiälä April 2018

From late night 14 April 2018 to morning 15 April 2018, an elevated aerosol layer between 2 km to 4 km a.g.l. was observed above Hyytiälä (Figure 11). The averaged profile from 03:00 to 04:00 UTC on 15 April 2018 shows an elevated aerosol layer with $\delta_{aerosol} = 0.24 \pm 0.008$, while in the boundary layer (from the surface to 1.5 km a.g.l.), $\delta_{aerosol} = 0.12 \pm 0.004$. This substantial difference in $\delta_{aerosol}$ indicates that the elevated aerosol had likely undergone long-range transport to the site. The FLEXPART simulation of this layer (Figure S5) indicates that the air mass was not in contact with the surface for 4.5 days prior to arrival over Hyytiälä. However, 5-7 days before arrival, a contribution from the surface layer was seen coming mostly from the western Sahara, with minor contributions from the Mediterranean (Figure S5). This, together with the high $\delta_{aerosol}$, suggests the layer to be mostly Saharan dust; the $\delta_{aerosol}$ value is comparable to values reported in the literature at other wavelengths (Freudenthaler et al., 2009; Groß et al., 2011; Burton et al., 2015; Haarig et al., 2017b). However, it is slightly lower than the dust value of 0.30 reported by Vakkari et al., (2021) at 1565nm. This could be due to the decrease of $\delta_{aerosol}$ in long range transport dust layer, resulted from gravitational settling of large dust particles during long range transport (Haarig et al., 2017b).
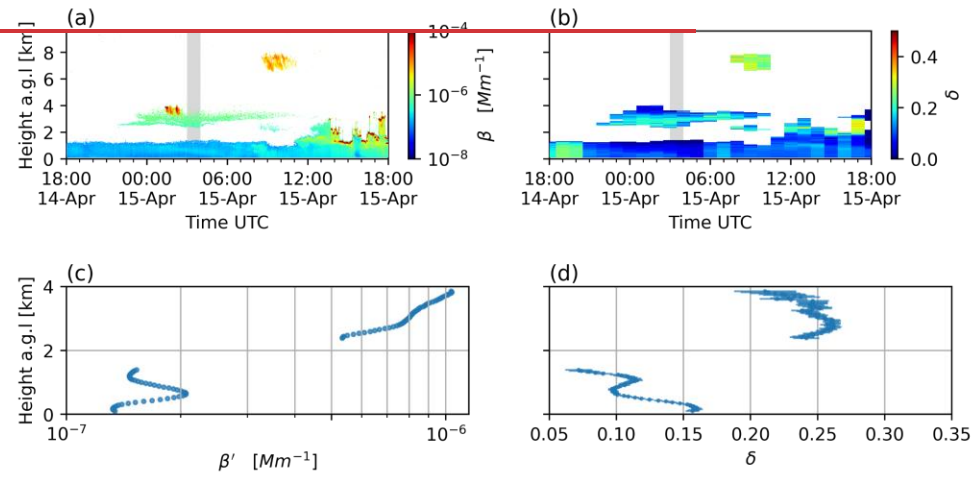

Figure 11: Profiles on 2018-04-14 to 2018-04-15 at Hyytiälä a) Attenuated backscatter (β'), b) 1-hour averaged depolarization ratio (δ). And 300m running mean (in shaded area) on 2018-04-15 03:00-04:00 UTC of c) β' profiles c) $\delta_{aerosol}$ profiles

3.5.2 Utö May 2017

On 13 May 2017, an elevated aerosol layer between 800 m a.g.l and 2 km a.g.l was observed over Utö (Figure 12). For the period on 2017-05-13 from 18:00 to 20:00 UTC, $\delta_{aerosol}$ of this elevated layer is at 0.23 ± 0.01 and decreases with height (Figure 12d and 12f). At the same time, $\delta_{aerosol}$ of the boundary layer aerosol from the surface up to 500 m is at 0.26 ± 0.009.
The air mass history shows that this layer originated from both offshore in the Baltic Sea and from continental Finland (Figure S6). The layer was elevated from the surface only 1 day before arrival and remained at a height of 1-2 km a.g.l. In this region there are no other known sources of aerosol with such high $\delta_{aerosol}$ except for pollen, and thus this layer is probably pollen transported from the surrounding continental areas. From 2017-05-14 00:00-05:00 UTC, this layer merged into the boundary layer, with the maximum value of $\delta_{aerosol}$ in the boundary increasing to 0.30 ± 0.007. The value of $\delta_{aerosol}$ is comparable to previous observations for spruce pollen in boreal forest; 0.269 ± 0.005 (Vakkari et al., 2021) and 0.29 ± 0.10 (Bohlmann et al., 2021).
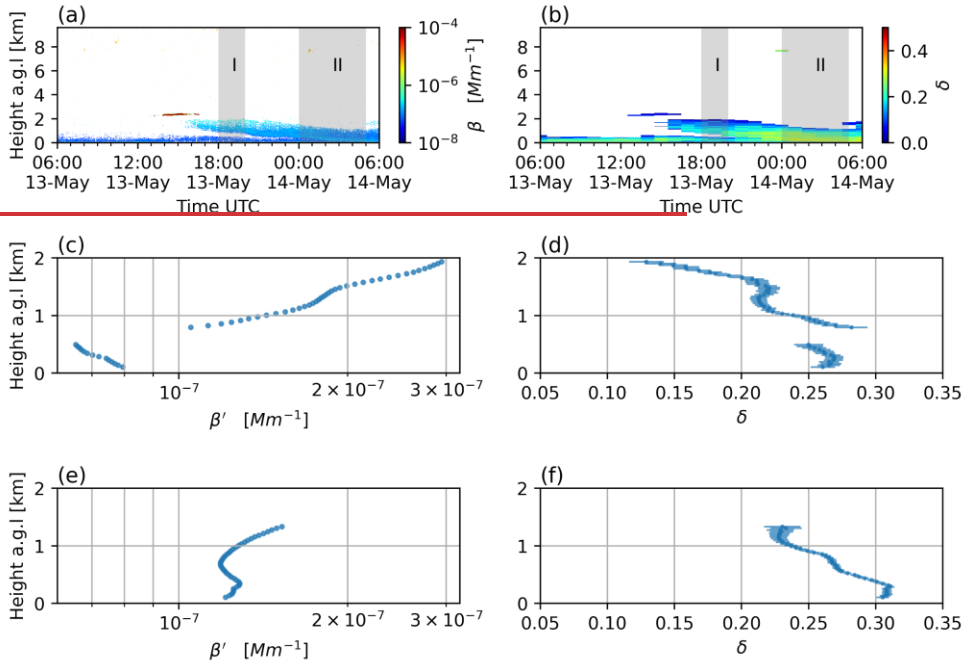
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Figure 12: Profiles on 2017-05-13 to 2017-05-14 at Utö a) Attenuated backscatter (β') b) 1-hour averaged depolarization ratio (δ). And 300m running mean (shaded area I) at 2017-05-13 18:00-20:00 UTC of c) β' profile c) δ_aerosol profile. 300m running mean (shaded area II) at 2017-05-14 00:00-05:00 UTC of e) β' profile c) δ_aerosol profile

4 Conclusions

In this study, the use of Halo Doppler lidars for long-term monitoring of the δ of aerosol δ_aerosol was investigated with a four-year long data set from the Finnish remote sensing network. The first aim was to investigate the stability of the instrumental noise floor and internal polarizer performance. The second aim was to characterise the seasonal and diurnal variation of the δ of aerosol δ_aerosol at a wavelength of 1565 nm at four different sites in Finland. In order to facilitate the second aim, an Aerosol Identification algorithm was created utilizing only data from Halo Doppler lidars.

The instrumental noise floor was assessed through the time series of the standard deviation of SNR_{co} in noise-only (aerosol- and hydrometeor- free) conditions. Four of the six systems included in this study presented a very stable noise floor, but two systems had up to 6% variability in the σ_{SNR} time series, even after changes in the configured integration time were taken into account. However, visual inspection did not indicate major issues in the data from the systems with increased variability in σ_{SNR}.

The time series of δ at liquid cloud base did not indicate long-term changes in the bleed-through of the Halo Doppler lidar internal polarizer. Overall, the observed values were comparable to previous case studies, though some elevated (δ > 0.02) values were observed especially during the wintertime, possibly due to the inclusion of some mixed-phase clouds erroneously classified as liquid-only. Also, the configuration with very long integration time of 27.5 s resulted more frequently in elevated δ at liquid cloud base, leading to a long tail in the otherwise Gaussian distribution of δ. Excluding the tail of the distribution resulted in bleed-through estimates that are very similar for all five instruments that were included in the study, ranging from

0.004 to 0.011. Including all the δ values would increase the bleed-through estimate at most from 0.008 ± 0.007 to 0.020 ± 0.022 for the Hyytiälä-46 lidar, for other systems the effect is smaller.

Two cases of elevated, long-range transported aerosol were studied in more detail. From late night 14 April 2018 to the morning 15 April 2018, an elevated aerosol layer between 2 km to 4 km a.g.l. was observed at Hyytiälä. The unusually high δ of aerosol ~~δaerosol~~ of 0.24 ± 0.008 for April at Hyytiälä for this event, together with aerosol subtype from CALIOP, dust aerosol optical depth from CAMS and air mass history analysis from FLEXPART indicate that this layer could be identified as Saharan dust~~indicating a source contribution from North-West Africa, mean that this layer could be identified as Saharan dust~~. The δ ~~δaerosol~~ value of this aerosol layer is comparable to previous observations of long-range transported Saharan dust (Freudenthaler et al., 2009; Groß et al., 2011; Burton et al., 2015; Haarig et al., 2017b; Vakkari et al., 2021). On 13 May 2017 in Utö, an elevated aerosol layer with ~~elevated δaerosol~~ high δ was observed between 800m and 2km a.g.l.~~.~~ The air mass footprint of this layer was from the Baltic Sea and surrounding areas, suggesting the layer contained pollen originating from continental areas and arriving at Utö at night. During this episode, the δ of aerosol ~~the δaerosol~~ ranged from 0.23 to 0.30, which was well within the range of previous reports for pollen in Finland (Bohlmann et al., 2019, 2021; Shang et al., 2020; Vakkari et al., 2021).

From the four-year observations of the δ of aerosol~~δaerosol~~, ~~it is~~we found that ~~δaerosol~~it is surprisingly similar at all four sites in Finland. The overall δ of aerosol ~~δaerosol~~ are 0.13 ± 0.08 in Hyytiälä, 0.11 ± 0.07 in Vehmasmäki, 0.09 ± 0.07 in Utö and 0.07 ± 0.08 in Sodankylä. All these sites have low value of δ of aerosol ~~δaerosol~~ in the winter months and higher ~~δaerosol~~ values in the summer months, which is attributed to the presence of irregular-shaped pollen particles in relatively clean background air. The highest monthly averages in the δ of aerosol ~~δaerosol~~ are observed in May and June. During this period, the δ of aerosol ~~δaerosol~~ has a strong diurnal cycle: at Hyytiälä, Vehmasmäki and Sodankylä, ~~δaerosol~~it peaks in the afternoon, while at Utö the peak is several hours later in the night. This difference for the island station of Utö is attributed to the time lag required for pollen to be transported from the mainland to the island. Additionally, the δ of aerosol ~~δaerosol~~ in the nearest 300 m a.g.l. was found to have a negative correlation with relative humidity at the surface. However, this negative correlation could stem from either hygroscopic growth or from concurrent changes in the aerosol type e.g., due to pollen release diurnal cycle. Attributing which factor plays a more dominant role is challenging and would require more profiling measurements.

~~Two cases of elevated, long-range transported aerosol were studied in more detail. From late night 14 April 2018 to the morning 15 April 2018, an elevated aerosol layer between 2 km to 4 km a.g.l. was observed at Hyytiälä. The unusually high δaerosol of 0.24 ± 0.008 for April at Hyytiälä for this event, together with air mass history analysis from FLEXPART indicating a source contribution from North-West Africa, mean that this layer could be identified as Saharan dust. The δaerosol value of this layer is comparable to previous observations of long-range transported Saharan dust (Freudenthaler et al., 2009; Groß et al., 2011; Burton et al., 2015; Haarig et al., 2017b; Vakkari et al., 2021). On 13 May 2017 in Utö, an elevated aerosol layer with elevated δaerosol was observed between 800m and 2km a.g.l. The air mass footprint of this layer was from the Baltic Sea and surrounding areas, suggesting the layer contained pollen originating from continental areas and arriving at Utö at night. During this episode the δaerosol ranged from 0.23 to 0.30, which was well within the range of previous reports for pollen in Finland (Bohlmann et al., 2019, 2021; Shang et al., 2020; Vakkari et al., 2021).~~

In conclusion, the long-term performance of Halo Doppler lidars was found to be satisfactory for the determination of the δ of aerosol~~δaerosol~~ for continuous monitoring purposes. Furthermore, the ~~aerosol~~ Aerosol ~~i~~Identification algorithm developed for this study, which is based on Doppler lidar only, was found to agree with the aerosol mask from the multi-instrument Cloudnet classification algorithm for more than 90% of time. This extends the capabilities of the Finnish remote sensing network (Hirsikko et al., 2014) in identifying potentially hazardous aerosol particles, which was one of the motivations in choosing Halo Doppler lidars for the network.

~~*Code availability*~~

~~The aerosol identification algorithm is available at https://github.com/vietle94/HaloDopplerLidar-AerosolIdentification.~~

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