1. Lidar CNR Uncertainties Calculation

In the calculation of the CNR uncertainties, we use the power spectrum, as recommended by ISO for radial wind speed assessment (ISO, 2017). Instead of using the power spectrum of radial wind speed, however, we use the power spectrum of CNR. Along the time axis, we calculate the power spectral density (PSD) for each range. An example is shown in Figure S1 to illustrate the method. From this spectrum, we calculate the uncertainty of CNR. At high frequencies, the power spectrum is dominated by a flat level due to noise (green line in Figure S1). The uncertainty σ is given by

$$\sigma = \sqrt{\Delta f * \varphi} \tag{S1}$$

where bandwidth Δf corresponds to the frequency region of the flat level (about 0.5 Hz in the example in Figure S1), and the average PSD value at the flat level is φ (0.1 dB^2/Hz in the example). Hence, in the example in Figure S1, CNR uncertainty σ_{CNR} at range, 2040 m was approximately 0.22 dB. Analyzing data from the two IMO lidars from two case studies, we found CNR uncertainties within aerosol layers of less than 1 dB in June 2019, and less than 0.5 dB in July/August 2019, respectively.

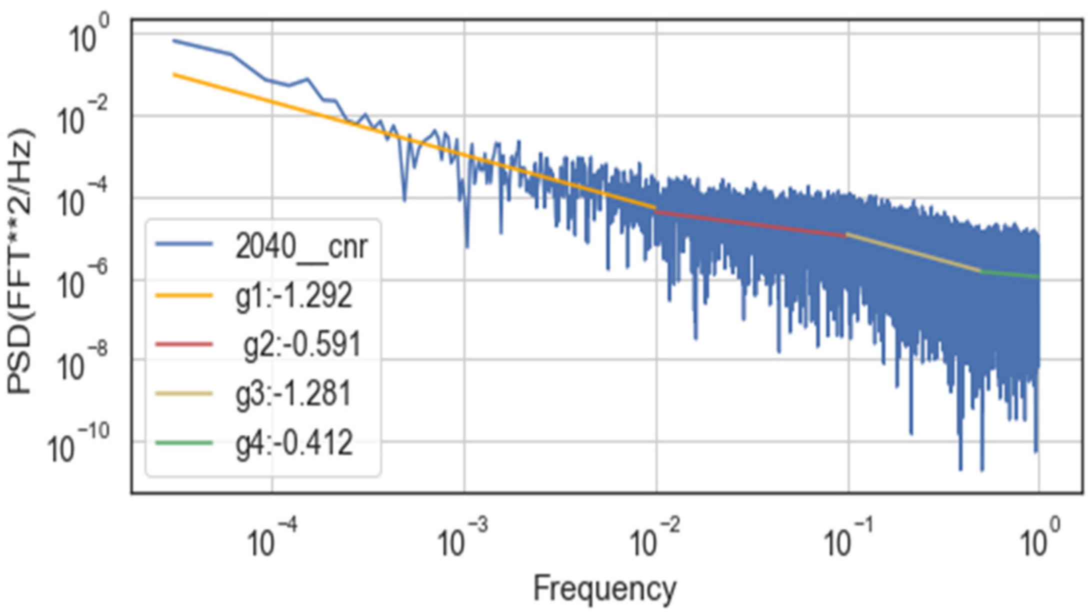

Figure S1. Log-log plot of CNR power spectrum along time within 24 h, at range 2040 m of a WindCube Scan 400S, using test data from Leosphere facilities. Coloured lines g1 to g4 mark the gradients of linear regression lines of different sections of the spectrum.

2. Lidar Data Processing: Relative Backscatter Coefficients Retrieval

The relative attenuated backscatter coefficient at a certain range gate x can be expressed as:

$$\beta_{relative}(x) = \beta_{calibration}(x) \times 10^{\left(\frac{CNR(x) - f_{dB}(x)}{10}\right)} \tag{S2}$$

where $\beta_{calibration}(x)$ is the calibrated backscatter coefficient, $CNR(x)$ is the CNR in dB at range x, and $f_{dB}(x)$ is the telescope function in the logarithmic scale (in dB). $\beta_{calibration}$ is a calibration value determined by several parameters:

$$\beta_{calibration}(x) = \beta_{ref} \times 10^{\left(\frac{f_{max} + \Delta CNR - CNR_{ref}}{10}\right)} \tag{S3}$$

CNR_{ref} and β_{ref} are reference values that were determined by the manufacturer during a calibration field campaign and directly link the CNR values to the relative backscatter coefficient. ΔCNR is an offset CNR value, depending on the pulse length or vertical resolution of lidar. In our case, we use 100 m resolution, corresponding to −3 dB. f_{max} is the maximum value of f_{dB}, which is the peak value at the focus point. The telescope function f(x), also called the Lorentzian curve fit function, is determined using data obtained during a calibration scan under homogeneous conditions. The fitting function is:

$$\mathrm{y(x)} = y_0 + \left(\frac{2A}{\pi}\right)\left(\frac{w_0}{4(x-x_0)^2 + {w_0}^2}\right) \quad \text{(S4)}$$

where y(x) is the CNR at range x in linear scale, x_0 is the focal length, y_0 is the noise floor, A is the amplitude above the noise floor, and w_0 is the full width at half maximum. Four fitting parameters (x_0, y_0, A, w_0) were calculated for each valid profile during the calibration scan, and the mean value of all fitting results are the parameters we used for the Lorentzian curve fit function:

$$f(x) = y_0 + \left(\frac{2A}{\pi}\right)\left(\frac{w_0}{4(x-x_0)^2 + {w_0}^2}\right) \quad \text{(S5)}$$

With this function, we are able to retrieve the relative backscatter coefficient $\beta_{relative}$ from CNR, using equation S2. This $\beta_{relative}$ is not only range corrected but also focus corrected, backscattered signal received by lidar. This calibration should be done whenever the lidar was moved or reset. The transmission term of the lidar equation is considered negligible, at least for near-field range gates. The focal correction is applied for all lidar backscatter coefficients (β) presented in this work.

3. Lidar Data Processing: Depolarization Ratio Retrieval

The scanning WindCube detects a total backscattered signal and the fraction of the signal that is perpendicularly polarized, relative to the polarization of the emitted beam. The lidars do not store received signals directly, but CNR in dB, which is the signal intensity on a logarithmic scale. In depolarization mode, the lidar will store $CNR_{\perp}$ (perpendicular) and CNR_{total}, which is the combination of $CNR_{\perp}$ and $CNR_{\parallel}$ (parallel), separately. Then depolarization ratio (δ) is defined by eq. S6 and eq. S7:

$$\delta = \frac{1}{1 + \mathrm{P}} \quad \text{(S6)}$$

where P indicates the power received by lidar parallel to the plane of polarization direction:

$$P = 10^{(CNR_{total} - CNR_{\perp})/10} \quad \text{(S7)}$$

The depolarization ratio ranges from 0 to 1: the value close to 0 means particles can be assumed to be spherical, which is the case for liquid droplets, such as cloud droplets. On the contrary, a larger value indicates the particles are non-spherical, which means they are more likely to be solid particles, such as ice, dust, or ash particles.

4. Lidar Data Processing: Data Screening

The lidar output is expected as one CNR_{total} profile followed by one $CNR_{\perp}$ profile when depolarization mode is turned on. However, due to the instrumental instability, sometimes there will be $CNR_{\perp}$ profiles missing. Figure S2 shows the time height cross-section of CNR (top panel) and δ (bottom panel) on July 28, 2019. The left panels (a and c) show the original data and the right panels (b and d) show the data screened by the lag time between profiles. Figure S2(a) shows the original CNR data on July 28, 2019, and vertical stripes reflect missing $CNR_{\perp}$ profiles. Consequently, we screen the data by checking the time difference between two consecutive CNR_{total} profiles. The time difference depends on the scan strategy, and in our case, it is around 2.7 seconds. We exclude all the profiles with corresponding time reference longer than 3 seconds, and the results are shown in Figure S2(b). In this way, most stripes are removed. Lidar calculate δ only when both $CNR_{\perp}$ and CNR_{total} profiles are available, thus the stripes removed from δ profiles. We will use the same data screening strategy for all lidar data for consistency, including the backscatter coefficient and depolarization ratio.

In some other cases, such as June 14 (Figure S3(c)), there are stripes when δ is high. This is observed in several cases and is caused by the change of the focal function parameter from one profile to another. Normally, the focus behaviour along the profile cancels out when calculating the depolarization ratio. However, as $CNR_{\perp}$ and CNR_{total} are measured subsequently and not simultaneously, profile-to-profile changes in the telescope function can cause the artifact is shown in Figure S3 (c). The vertical stripes change colour around the focal length of about 700 m. This happens when the assumption of a constant telescope function from one profile to the next is not met. Then, δ can be underestimated below the focal point and overestimated above, or vice versa. With 15 minutes averaging, most stripes can be smoothed, and a qualitative analysis of δ is possible. In the same case, β is increasing with altitude in the noise regime. To remove the noise, we filtered the lidar data when the CNR reaches a threshold. Boquet *et al.* (2016) measured the CNR threshold for a WindCube 200s with 100 m resolution as −28.6 dB. Considering the atmosphere in Iceland is clean, we use −28 dB in this study. Figure S3(d) shows the temporally smoothed and CNR filtered results.

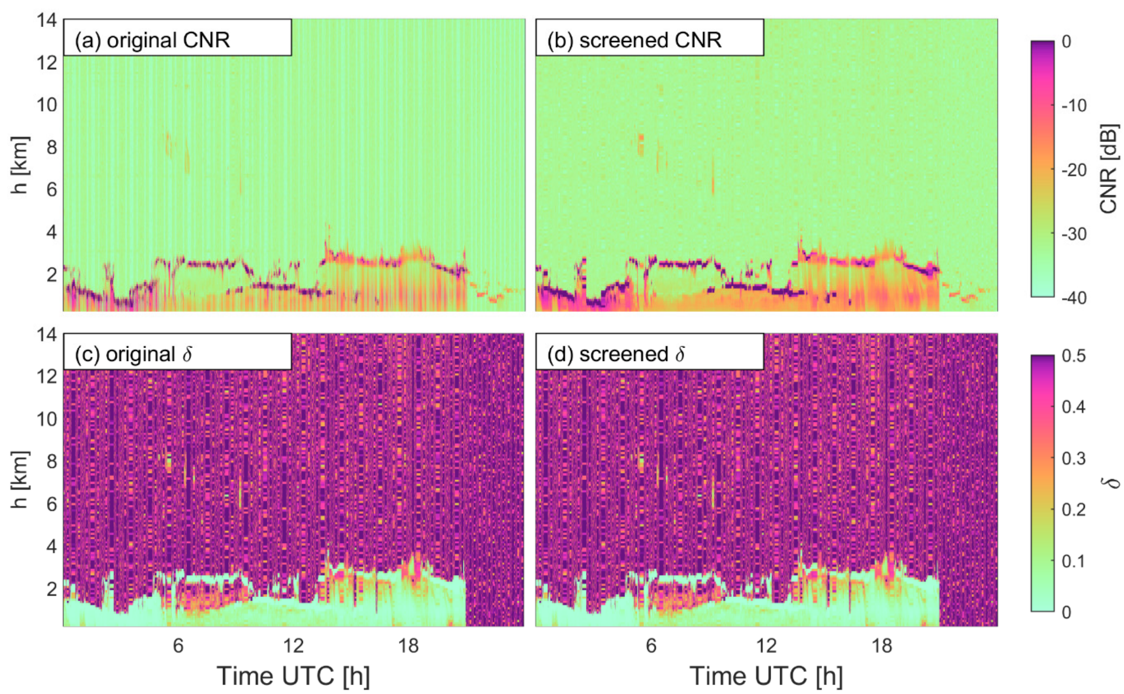

Figure S2. Time height cross-section of CNR (top panel) and δ (bottom panel) on 2019 June 28. The left panels (**a** and **c**) show the original data and the right panels (**b** and **d**) show the data screened by the lag time between profiles.

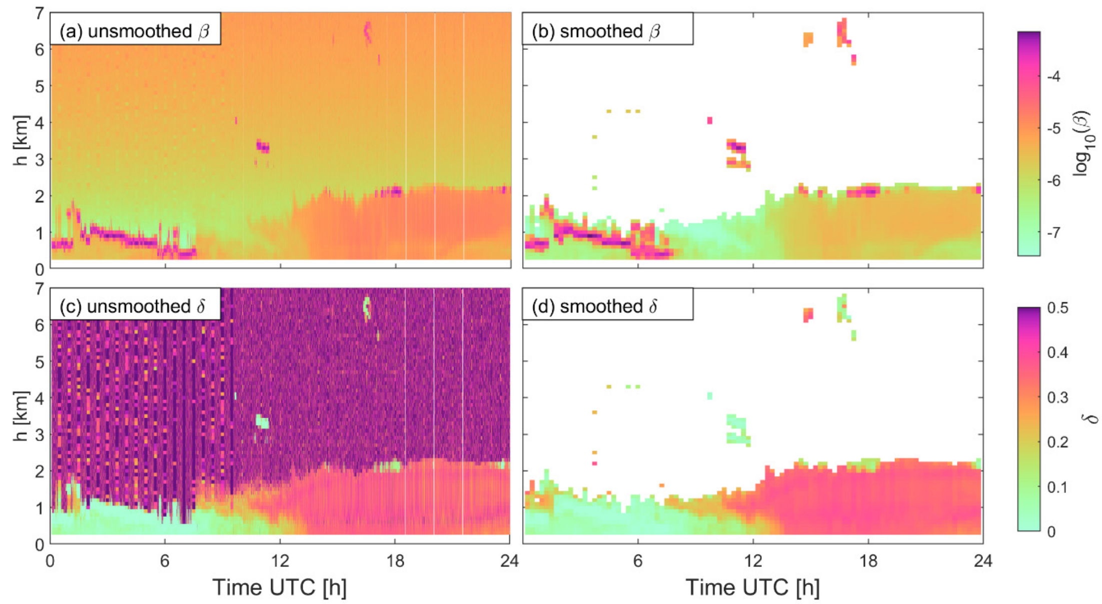

Figure S3. Time height cross-section of β (top panel) and δ (bottom panel) on 2019 June 14. The left panels (**a** and **c**) show the unsmoothed data and the right panel (**b** and **d**) shows the smoothed data with a 15 minutes average, and data points lower than −28 dB were removed.

5. Ceilometer Dark Measurement

The dark measurement, or termination hood measurement, can be done by covering the window of ceilometer using a termination hood. In this way, the entire emitted signal is attenuated, and the received signal can be attributed entirely to the background signal. In this study, we have performed two dark measurements with the ceilometer on the trailer (CL31): one is on December 3rd, 2018 and the other is on June 6th, 2019. Both measurements lasted around one day (~22 hours and 26 hours). In our study, we checked the signal at the 5th range gate (50 to 60 m) and use the profile when it reaches a stable characteristic value as the beginning of the dark measurement.

6. Factor C and Cloud Base Height

The factor C used for ceilometer data correction is calculated as the mean value of factor C when there is no cloud and factor C is relatively stable, see the Figure S4 below.

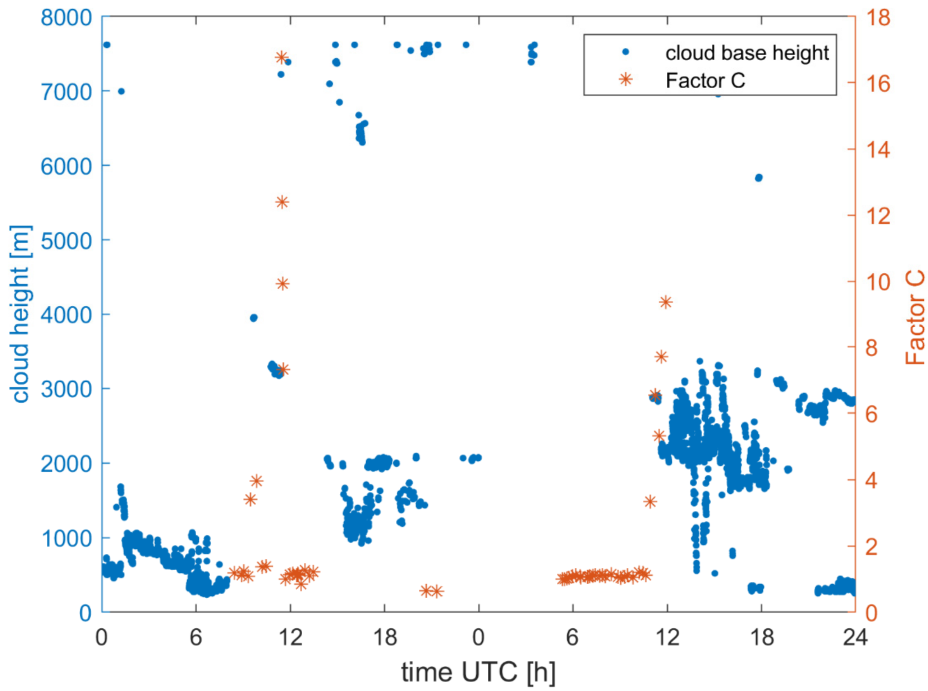

Figure S4. Cloud base height (blue) measured by ceilometer and factor C (orange) calculated based on ceilometer and sun-photometer measurements on June 14 and 15, 2019, at Reykjavik. The time axis starts from 00:00 UTC, June 14, 2019, covering two days.

7. Datasets Available Online

Some datasets are available online on the following websites.

Sun-photometer data from AERONET: https://aeronet.gsfc.nasa.gov/. Last accessed in September 2020.

PM10 and PM2.5 concentration measurements from the Environment Agency of Iceland: https://loftgaedi.is. Last accessed in December 2019. The instrument used for this study was BAM 1020 Continuous Particulate Monitor, the specifications can be found at https://metone.com/products/bam-1020/. Last accessed in November 2020.

Radio sounding measurements from at the University of Wyoming's website: http://weather.uwyo.edu/upperair/sounding.html. Last accessed in March 2020.

HYSPLIT backward trajectory simulation: https://www.ready.noaa.gov/HYSPLIT.php. Last accessed in August 2020.

8. Retrieval of Ceilometer Extinction Coefficient Profile

The backscatter signal measured by lidar/ceilometer at a certain range P(r) is expressed as:

$$P(r) = \frac{C_L}{r^2} \beta \mathrm{T}^2 \tag{S8}$$

where C_L is the lidar constant, assumed as 1 in this study; r is the range gate, β is the total backscatter coefficient and T^2 is the transmission term. The scattering of light in the atmosphere differs with the size of scatterers relative to the wavelength: if the scatterers are significantly smaller than the wavelength, the Rayleigh theory can be applied. If this is not the case the Mie theory has to be applied. In this study we treat the scatterers from aerosol particles (p) and molecules (m) separately:

$$\beta = \beta_p + \beta_m \tag{S9}$$

$$\alpha = \alpha_p + \alpha_m \tag{S10}$$

The particle lidar ratio $S_p = \frac{\alpha_p}{\beta_p}$ is assumed as a height independent value, in this case we use $S_p = 55\ sr$ (Ansmann *et al.*, 2011).

After the correction of ceilometer RCS profiles (see example in Figure 2, left), we can apply the backward Klett inversion algorithm on the corrected RCS profile to retrieve the particle backscatter coefficient profile $\beta_p(r)$, and the extinction coefficient profile $\alpha_p(r) = \beta_p(r)S_p$. It also follows the conditions that the integrated extinction coefficient from ceilometer data agrees with AOD measured from sun-photometer (Wiegner & Geiß, 2012).