

Painting a More Colorful Picture of the Atmosphere:

Hyperspectral Lidar for Characterization of Atmospheric Constituents

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Abstract

Atmospheric particles play a significant role in radiative forcing through light scattering and new methods are called for to better characterize their impact. Typical Lidar systems emit single wavelength light, not utilizing full spectral information from scattering measurements.

This thesis aims to use a hyperspectral lidar system to measure the spectral dependence of scattering and absorption of different atmospheric constituents. This was done by emitting broad band short-wave infrared light and collecting the backscattered light on a 2D InGaAs detector, mapping wavelength on one axis and range on the other.

The system was tested along a horizontal path over a range of 700 meters, where ambient measurements were made under different atmospheric conditions as well as controlled releases of aerosol plumes. The system successfully retrieved hyperspectral signals from multiple atmospheric constituents which revealed spectral features in absorption and scattering. From these measurements, wavelength dependent backscatter and attenuation coefficients could be calculated.

The study demonstrates the capability of the hyperspectral lidar to characterize particulate matter for a broadband spectrum, which can provide information on microphysical properties. The broad applicability of the method showcased the possibility to measure simultaneous backscatter and absorption for different spectral bands, supporting future applications in atmospheric monitoring studies.

Contents

1	Introduction	1
1.1	Aerosols, Lidars, and the inverse problem	1
1.2	Hyperspectral Lidar	2
2	Theory / Background	3
2.1	Aerosols and Climate	3
2.2	Lorenz-Mie Theory	4
2.3	Elastic Backscatter Lidar	6
2.4	Scheimpflug Lidar	8
2.5	Lidar Ratio and Extinction coefficients	8
2.6	Inversion and ill-posedness	10
2.6.1	Bayesian inference model	11
2.7	Scattering of larger atmospheric targets	12
2.8	Molecular absorption and DIAL	12
3	Methodology	15
3.1	Setup	15
3.1.1	Supercontinuum Source	15
3.1.2	Receiver	15
3.1.3	Line of Sight	15
3.2	Measurement Principle	17
3.3	Data Acquisition	18
3.4	Pre-processesing and Calibration	18
3.5	Experimental measurements	21
3.5.1	Hydrometeros	21
3.5.2	Ambient measurements	21
3.5.3	Plumes	21
3.6	Signal Analysis	22
3.6.1	Extinction	22
3.7	Backscatter	23
3.8	Lidar ratio	23
3.9	DIAL	23

3.10 Simulations	24
4 Results	26
4.1 Hydrometeors	26
4.1.1 Fog Clouds	26
4.1.2 Snow	29
4.2 Plumes	29
4.3 Ambient aerosols	35
4.4 Water Vapour and DIAL	35
4.5 Retrieval possibilities	39
5 Discussion	43
5.1 Spectral response of atmospheric targets	43
5.2 Intepretation	43
5.3 Limitations	44
5.3.1 Optical Depth and Multiple Scattering	44
5.3.2 Particle distributions and spectral dependence	44
5.3.3 Signal to noise	45
5.3.4 Retrieval and calibration limitations	45
5.4 Evaluation	45
5.5 Improvements	46
6 Outlook	47

1 Introduction

1.1 Aerosols, Lidars, and the inverse problem

Aerosols are liquid or solid particles, fine enough to be suspended in the air for extended periods of time. This broad term thus encapsulates soot emitted from a chimney, sand swept from the Sahara, pollen blown from a tree and salt spouted from the sea. These tiny particles have a significant impact on Earth, making them important to study. Aerosols in the lower troposphere pose health problems through inhalation. At higher altitudes, they have another important effect which has to do with their ability to interfere with sunlight. Greenhouse gases are known to heat the planet by absorbing thermal radiation from the Earth while aerosols primarily affect the climate through scattering sunlight. Their influence or radiative forcing, as compared to greenhouse gases however, remains uncertain.

As the particles in question are rather inaccessible to measure directly, remote sensing techniques have become prominent in identifying them at a distance. Sun photometers measure the effect of particles on transmitted sunlight through the atmosphere. Lidars (light detection and ranging) on the other hand, are instruments that can emit their own light to probe atmospheric constituents. Lidars are flexible and can be positioned both on the ground for local measurements and in space to scan the entire Earth.

The simplest atmospheric Lidars operate by emitting monochromatic light and measuring the portion of it that returns at the same frequency, commonly referred to as elastic backscatter. These are thus referred to as elastic backscatter Lidars. The distance to the particles is calculated from the time it takes light to make a round trip. What remains is to classify and type the different aerosols at multiple distances. This requires knowledge of how different particles scatter light, which is described by the Lorenz-Mie theory.

Simplifying the problem by assuming the aerosol particle is a homogeneous sphere, Lorenz-Mie theory predicts that the three properties that uniquely characterise aerosol light scattering are its size and its real and imaginary refractive indices. One can then use this information to solve the inverse problem: determining the aerosol size and refractive index from the backscattered light. This is often the relevant question since we want to determine which particles make up our atmosphere.

The main issue with this approach is that, from a single observable, multiple variables must be determined. This is known as an ill-posed problem where the known quantities do not constrain the problem enough to obtain a unique solution. The difficulty can be mitigated by measuring, one way or another, the attenuation of the beam throughout the probed air column, i.e. the extinction. Furthermore, one can measure the backscatter and extinction at different wavelengths, adding more constraints. The problem remains, nonetheless, that the way particles mix, their size distribution, and how well they fit our Lorenz-Mie assumptions add more variables to solve for. The logical question is whether there is a solution to this

seemingly hopeless task. In this thesis, I propose that we broaden our view, by employing multiple wavelengths in the form of a supercontinuum.

1.2 Hyperspectral Lidar

Using a supercontinuum light source in the short-wave infrared wavelength regime, we hope to obtain the complete spectral dependence of elastically backscattered light from aerosols in a hyperspectral Lidar. Instead of a traditional setup that uses time-of-flight to calculate distance and a scanning technique to separate wavelengths, a purely geometric setup based on the Scheimpflug principle is used [1]. This allows one to obtain a focused depth of field which is spectrally resolved. This novel approach uses a set-up that is both compact and cost-efficient. From the retrieved data, one can strive to obtain a spectral fingerprint for every set of particles in the atmosphere.

The instrument used was originally designed for insect monitoring [2], where its ability to detect spectral signatures in specular reflection from wings at 90 m distance was demonstrated in determining wing thicknesses. In this thesis, the aim is to measure the spectral dependence of Lorenz-Mie scattering from aerosols at a range of up to 700 m as well as other spectral signatures. From that, determine physical quantities from the measured particles.

The goals of the thesis are the following :

1. Calibrate, Align and optimise Hyperspectral Lidar setup to maximise performance
2. Assess system performance for ambient aerosols as well as controlled releases of aerosol plumes
3. Evaluate spectral features of backscattered signals
4. Determine backscatter, extinction coefficients and Lidar ratios
5. Determine water vapour presence from absorption DIAL (differential absorption Lidar)

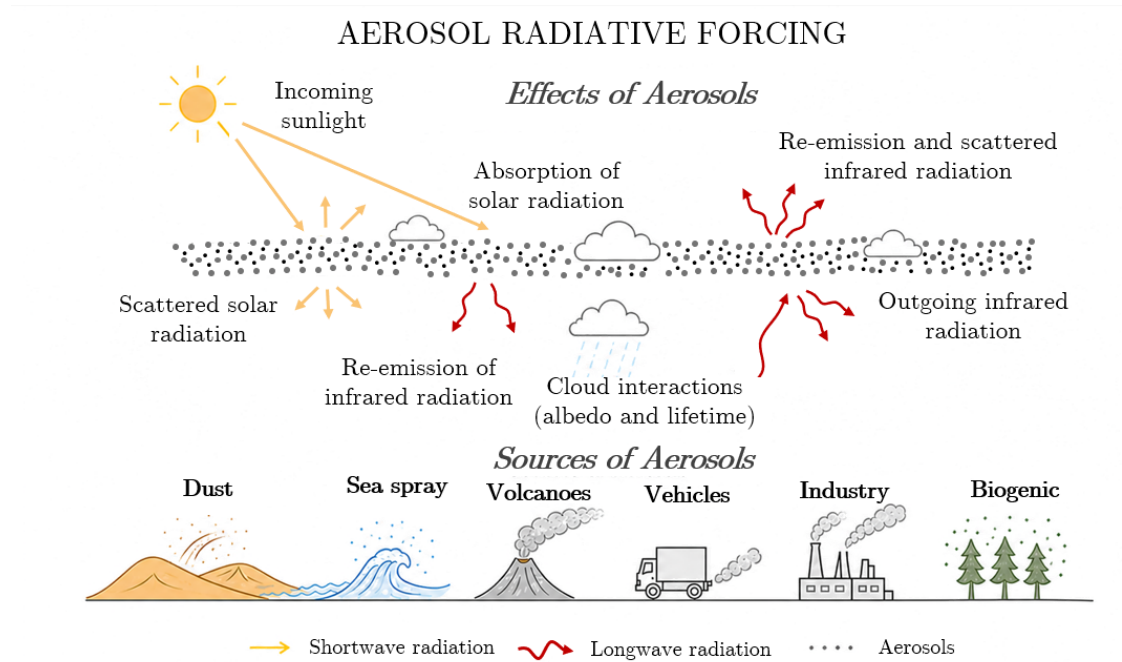


Figure 1: Aerosol scattering and absorbing effects together with sources.

2 Theory / Background

2.1 Aerosols and Climate

Much effort has been put into tracking the effects of climate change in recent years. Climate change is governed by shifts in the radiative energy balance of Earth, and processes where different droplets, particles and molecules scatter, absorb and emit light. The measure of the heating and cooling of the Earth is known as radiative forcing and is defined as the net radiative flux at the top of the atmosphere, measured in W/m^2 [3].

A large part of the positive radiative forcing is the greenhouse effect, which occurs due to thermal radiation from the earth being absorbed by gases in the atmosphere such as carbon dioxide and water vapour, and then re-emitted in all directions. This has a heating effect on the climate and has been extensively studied and quantified. Gas concentrations and absorption spectra in the atmosphere are known and therefore their radiative forcing is well-constrained [4].

Liquid and solid particles also scatter light and naturally have an effect on the climate as well. Apart from interacting with light directly, they also have indirect effects, such as acting as cloud condensation nuclei. In Figure 1, radiative forcing of aerosols is shown together with different sources. It highlights how both absorption and scattering of sunlight and outgoing infrared radiation affect radiative forcing. Aerosols are typically good scatterers and generally have a negative radiative forcing, cooling the climate [4].

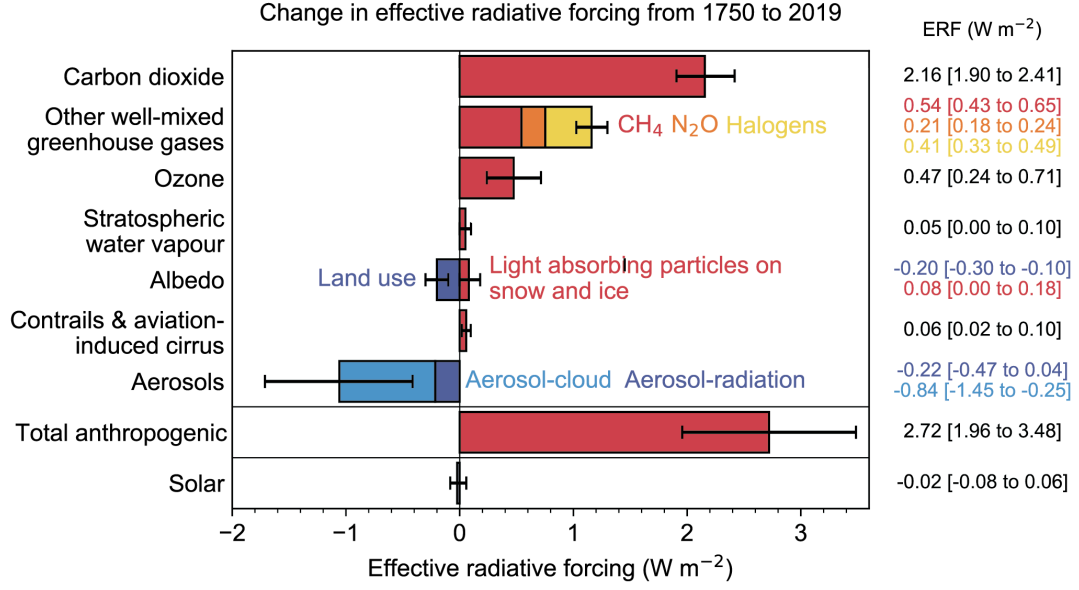


Figure 2: Radiative Forcing of different sources, source IPCC, 2021: Chapter 7 [4].

The effects of aerosols on the climate have so far been difficult to quantify and there are large uncertainties on the effect of aerosols on the climate, highlighted in Figure 2.

Aerosols are typically emitted into the atmosphere by anthropogenic or natural causes where they can form from precursor gases that cluster together to become 0.1 to 1 micrometers in size. Larger, shorter-lived particles are also present, originating from wind action. Aerosols are empirically assumed to be distributed log-normally in size [5]:

$$\frac{dN(R)}{dR} = \frac{N}{\sqrt{2\pi R \log \sigma}} \exp \left[-\frac{1}{2} \left(\frac{\log R - \log R_{mod}}{\log \sigma} \right)^2 \right], \quad (1)$$

where R_{mod} is the mode radius, σ is the width and N is the total particle number density.

2.2 Lorenz-Mie Theory

The scattering properties of aerosols is described by Lorenz-Mie theory or simply Mie theory, which is the theory of how electromagnetic waves interact with particles. It assumes that the particle is a homogeneous sphere and scatters light only elastically [6]. The theory is based in the solutions to Maxwell's equations and is completely determined by the size of the particle and its complex refractive index. The result is that there is one scattering and one absorbing contribution of the particle to the electromagnetic field. The analytical form of the solutions can be computed and from this, measurable quantities can be derived. Two common ones

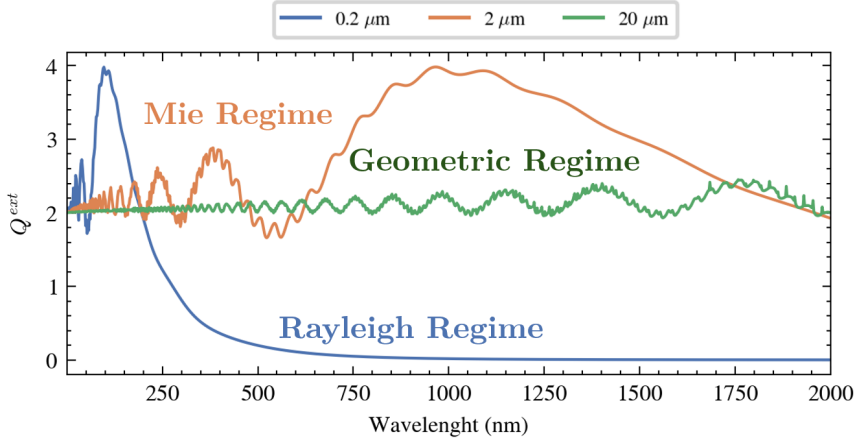


Figure 3: Extinction efficiencies for three different particle size with refractive index 1.33.

are the scattering cross section C^s which measures the effective scattering area of a particle, and the extinction cross section C^{ext} which is the scattering cross section plus the absorption cross section. This describes the effective area of the particle based on how much energy it removes from an incident electromagnetic wave. The analytic form of the scattering and extinction cross sections are:

$$C^s = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) (|a_n|^2 + |b_n|^2), \quad (2)$$

$$C^{ext} = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \Re(a_n + b_n). \quad (3)$$

Here, k is the wave vector, $\Re$ is the real part of the expansion coefficients a_n and b_n for the scattered electric and magnetic fields. In Figure 3, extinction cross sections are shown for particles of different sizes. The figure highlights three different regimes which are determined by the size parameter ($x = r/\lambda$). $x \gg 1$ is the geometric regime where the extinction cross section approaches a constant equal twice the geometric area of the particle. For $x < 1$, Mie scattering simplifies to Rayleigh scattering typical for atmospheric gases with a strong wavelength dependent decrease $\propto \lambda^{-4}$. For $x \approx 1$, extinction is highly wavelength-dependent, exhibiting two types of oscillatory behaviours: the first is a typical interference structure attributed to interference between the incident and forward-scattered waves. The second is a finer oscillatory structure called ripple structure, which is due to vanishing denominators in the expansion coefficients. This domain is called the Mie regime and is most applicable for Mie theory [6]. As the extinction cross section is highly dependent on wavelength in this regime, its form can be used to determine the scattering properties of different particles.

For Lidar, what is measured is only the portion of the scattered light that returns at the same angle it is emitted, or the backscattered light. Another useful quantity is thus the backscattering cross section C^{back} :

$$C^{back} = \frac{2\pi}{k^2} \left| \sum_{n=1}^{\infty} (2n+1)(-1)^2(a_n - b_n) \right|^2. \quad (4)$$

2.3 Elastic Backscatter Lidar

The elastic backscatter Lidar operates by emitting a beam of light at certain wavelengths that scatters elastically from the particles it encounters. It then measures the backscattered light which has been attenuated by extinction. The goal is then to relate the measured light to the microscopic scattering properties C^{ext} , C^{back} in order to retrieve information about the particles.

The microscopic scattering of particles from Mie theory are related to bulk, atmospheric observables through the following equations [7]:

$$\beta(r, \lambda) = \int_0^\infty \frac{C^b(R, \lambda, \Omega = 180)}{4\pi} n(R, r) dR, \quad (5)$$

$$\alpha(r, \lambda) = \int_0^\infty C^{ext}(R, \lambda) n(R, r) dR. \quad (6)$$

$\beta(r, \lambda)$ is the backscatter coefficient and measures the backscatter capability ($\text{m}^{-1}\text{sr}^{-1}$) of a particle distribution with number size density $n(R)$. $\alpha(r, \lambda)$ is the extinction coefficient and measures the portion of the beam scattered and absorbed per unit distance, measured in m^{-1} .

As the light from the Lidar interacts with particles, it is continuously attenuated, leading to an exponential decay. This follows from the Beer-Lambert law, where the removed light dI is proportional to the signal I , α , and distance x [8]:

$$-dI = \alpha I dx \implies I(x) = I_0 \exp\left(-\int_0^x \alpha(x') dx'\right) \quad (7)$$

β and α contributions to the Lidar signal are shown in Figure 4 (left) where the measured power is proportional to the product of the backscatter coefficient and the attenuation. The integrated extinction over the measured distance is known as optical depth (OD) $\int_0^r \alpha(r', \lambda) dr'$. The factor of 2 appearing in the exponential refers to the two-way transmission made by the light away and back to the instrument.

The full signal $P(r)$ of the Lidar at range r is governed by the Lidar equation [8]:

$$P(r, \lambda) = P_0 C \frac{\beta(r, \lambda)}{r^2} \Delta r \exp\left(-2 \int_0^r \alpha(r', \lambda) dr'\right), \quad (8)$$

where P_0 is the emitted power from the light source, C is an instrumental constant encapsulating different parameters such as conversion efficiency, receiver area, as well as the overlap between receiver FOV and light beam which is assumed to be one. The $1/r^2$ factor arises from spreading of the scattered light. The portion of the

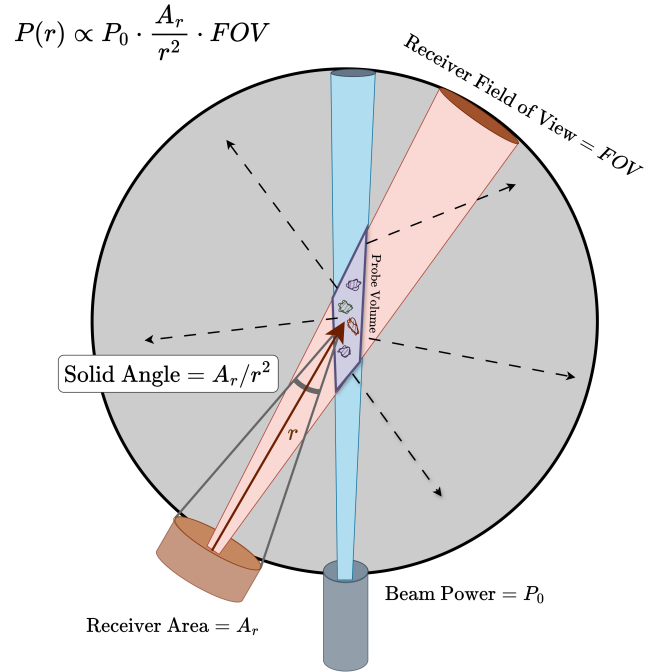
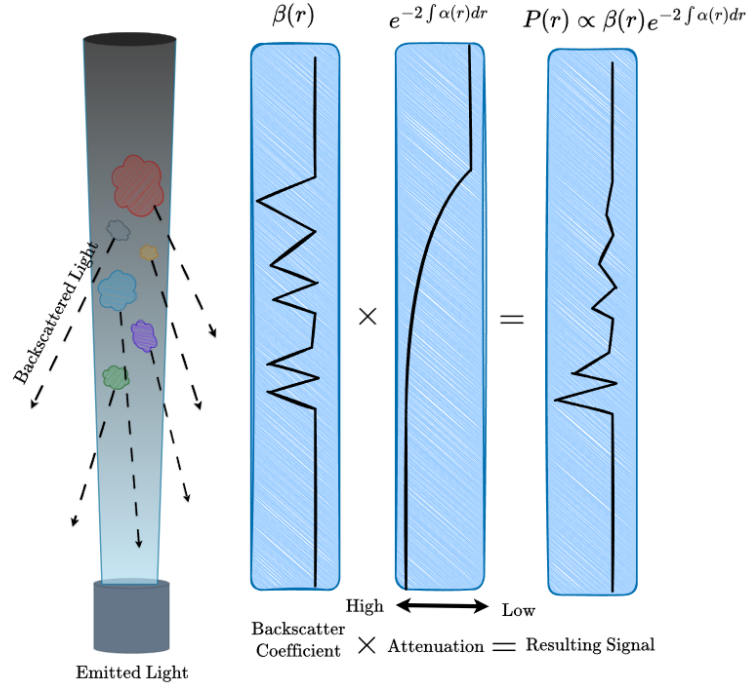


Figure 4: Heuristic argument of Lidar signal. Aerosol scattering light resulting in backscatter signal and attenuation (top). Lidar signal as a function of instrument setup (bottom).

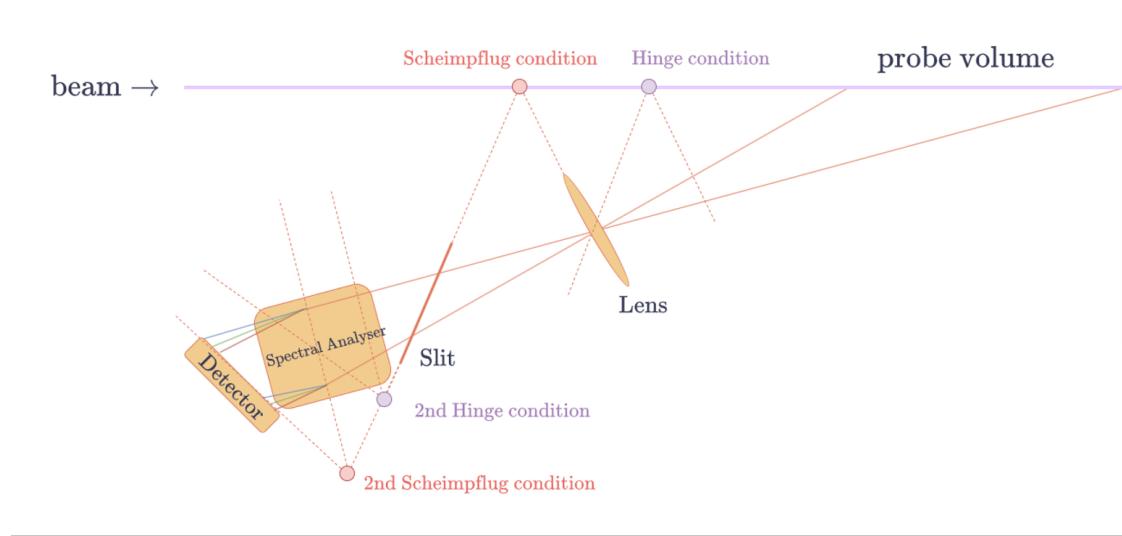


Figure 5: Scheimpflug and Hinge conditions applied twice to obtain a focused depth of field and spectral resolution

light that reaches the detector is then proportional to the solid angle subtended by the receiver area A_r/r^2 . The geometric factors of the Lidar equation are highlighted in Fig 4 (right), which also shows how the receiver Field of View (FOV) determines the size of the volume that can be measured [7].

2.4 Scheimpflug Lidar

What has not been mentioned from eq. 8 is the factor Δr which is the size of the range bins of the Lidar. Typically, range is measured using time-of-flight, where distance is determined from travel time of the emitted laser pulse, requiring high temporal resolution [8]. In this thesis, a geometric setup is used instead. This Lidar uses the Scheimpflug and hinge condition through a tilted sensor to obtain a focus throughout the entire field-of-depth as shown in Figure 5. Since each part of the beam is at a different angle from the detector, triangulation can be used to resolve distance. Here, the conditions are applied twice in order to resolve the wavelengths as well using a 2d dedector.

In Figure 6, it can be seen that due to the triangulation method, the size of the range bins increases with distance. This has the benefit of increasing the signal strength as Δr increases at large ranges to counteract the decreasing strength typically found as a result of the $1/r^2$ factor in eq 8. The tradeoff is that the resolution decreases, which is also affected by the beam width [9].

2.5 Lidar Ratio and Extinction coefficients

In order to retrieve useful quantities from atmospheric constituents, one needs to solve for the observables, α and β in the Lidar equation. The problem is that there is only one known variable: the received Lidar Power, and two unknowns. Rather,

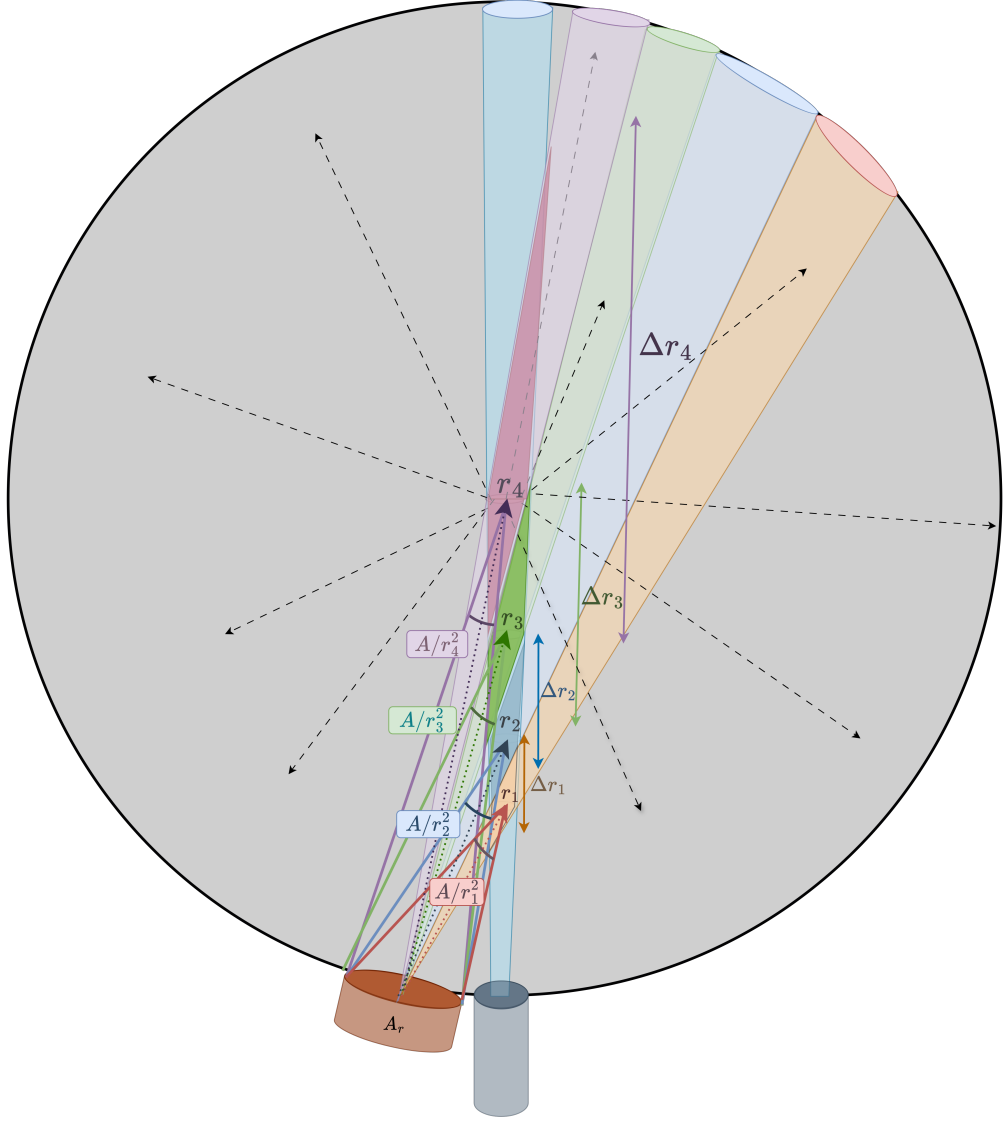


Figure 6: Sketch of different range bins probed by the Scheimpflug Lidar, highlighting the increase in bin sizes as distance increases.

it is four unknowns since the backscatter and extinction coefficient consist of two parts, molecular and aerosol contributions:

$$\beta = \beta_a + \beta_m, \quad (9)$$

$$\alpha = \alpha_a + \alpha_m$$

the molecular contribution to the lidar signal can be determined from atmospheric conditions [10], [8]. This again reduces the unknowns to two.

For an elastic Lidar, there are few ways of solving this issue, one of them are assuming the so called Lidar ratio α/β which allows the solution to be determined using a technique known as Klett-Fernald inversion [11]. Since the signal is proportional to the backscatter coefficient and decays exponentially through the extinction coefficient, one can use this relationship together with a boundary condition with known atmospheric qualities to determine the backscatter and hence the extinction along the path. Another option is to use the slope method [12] which assumes a homogeneous atmosphere such that the extinction coefficient can be determined from the exponential decay of the Lidar signal. Assuming a homogeneous atmosphere with constant α and β , the Lidar equation can be transformed into the following form:

$$\ln \left(\frac{Pr^2}{\Delta r} \right) = \ln(P_0 C \beta) - 2\alpha r, \quad (10)$$

taking the derivative we then get,

$$\alpha = -\frac{1}{2} \frac{d}{dr} \ln \left(\frac{Pr^2}{\Delta r} \right) \quad (11)$$

There are two types of Lidar which can determine extinction coefficient independently of backscatter. These are the HSRL (High spectral resolution Lidar) and Raman Lidars. The former measures the doppler broadening of molecular contribution of the lidar signal along the path to determine extinction coefficient. The Raman Lidar determines the inelastic scattering from molecules to determine the extinction [8]. These instruments are valuable to independently solve the Lidar equation while their drawbacks are low signal strengths.

2.6 Inversion and ill-posedness

Once the backscatter and extinction coefficient are determined, one can invert equations 5 and 6 to determine the complex refractive index and size distributions of different aerosols. However, since β, α are weighed sums of the size distributions and cross sections of different particles, the effect is smoothing and can be seen in the normalized extinction coefficient in Figure 7. This makes the problem more difficult.

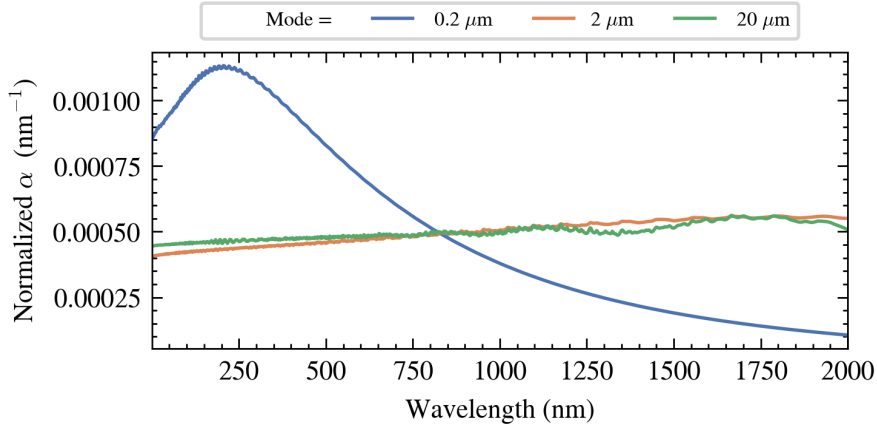


Figure 7: Extinction coefficients of 3 different size distributions with $\sigma = 2$ microns.

There is an inherent problem with the inversion of equations 5, 6, which is that they are ill-posed. In this case, the solutions suffer from being non-unique in that many types particle size distribution can yield equal backscatter and extinction coefficients. The second problem is that the solutions are unstable, making them highly sensitive to noise[13].

This is usually solved by imposing certain constraints on the problem to make the inversion well-behaved, one common example is Thikonov regularization [14]. Another approach is using a parametric optimization model, which assumes the form of the solution and minimizes the error by changing parameters. The one used in this thesis is a Bayesian inference model.

2.6.1 Bayesian inference model

Bayesian inference uses an algorithmic method to sample parameters and evaluate the most most probabilistic outcome. In this case, parameters of size distribution and refractive index are sampled to determine which fit most with measured α and β . The algorithm is based on Bayes Theorem[15]:

$$P(\theta|y) = \frac{P(y|\theta) \cdot P(\theta)}{P(y)} \quad (12)$$

which updates the posterior probability $P(\theta|y)$ of the parameters θ given the data y , based on the likelihood $P(y|\theta)$ of the data given the parameters. It also depends on the prior probability $P(\theta)$ associated with the parameters and the evidence $P(y)$ of the data being correct.

Assuming the measured data y_i has gaussian noise σ_i , the difference between y_i and hypothetical data from the parameters $f_i(\theta)$ is also distributed normally as:

$$p(y_i|\theta) = \frac{1}{\sqrt{2\pi\sigma_i^2}} \exp\left(-\frac{(y_i - f_i(\theta))^2}{2\sigma_i^2}\right), \quad (13)$$

Taking the logarithm and summing over all y_i , the log-likelihood probability density is:

$$\log p(y|\theta) = \frac{-1}{2} \sum_i \frac{(y_i - f_i(\theta))^2}{\sigma_i^2} + \text{constants} \quad (14)$$

The log-posterior probability density is then:

$$\implies \log p(\theta|y) = \frac{-1}{2} \sum_i \frac{(y_i - f_i(\theta))^2}{\sigma_i^2} + \log(p_i(\theta)) + \text{constants}.$$

Since the algorithm compares relative posteriors, the constants can be omitted.

2.7 Scattering of larger atmospheric targets

As extinction increases, inverting the Lidar equation becomes increasingly difficult. As aerosols group together, their number concentration increases leading to a larger extinction coefficient which can be seen in eq. (6). This increases the probability of photons interacting with particles in its path. At sufficiently high concentrations, photons may undergo multiple scattering events before reaching the detector [16]. This violates the single-scattering assumption of the Lidar equation that light is only scattered directly from particles in the beam path.

Multiple scattering is common in fog, clouds, rain and snow. Cloud droplets typically scatter light in the Mie and geometric regime, producing strong backscatter and extinction. Despite multiple scattering limitations, it is possible to determine microphysical properties from individual droplets and multi wavelength approach reduces the retrieval uncertainties [17].

Ice clouds are more complex in their scattering due to their composition of larger, non-spheric ice crystals[18]. Unlike spherical droplets, these particles exhibit more complex spectral signatures that cannot be fully described by classical Mie scattering alone. Due to their flatness and shape, they produce specular reflection and dispersive effects, resulting in different colors[19]. Depending on the shape, size and thickness, different colors may appear at different angles. This wavelength-dependency highlights the potential advantage of hyperspectral lidar measurements for characterizing these ice crystals.

2.8 Molecular absorption and DIAL

To utilize the full potential of a hyperspectral Lidar, we need to focus not only on scattering but also on absorption. This is where molecules enter the picture. In fact, investigating the solar irradiance on Earth in the SWIR range, (Figure 8), the largest features arise from two absorption bands of water. There are two ranges of absorption, one from around 1120 to 1160 nm and the strongest situated between 1350 and 1450 nm. These are the result of overtones in the vibrational bands of

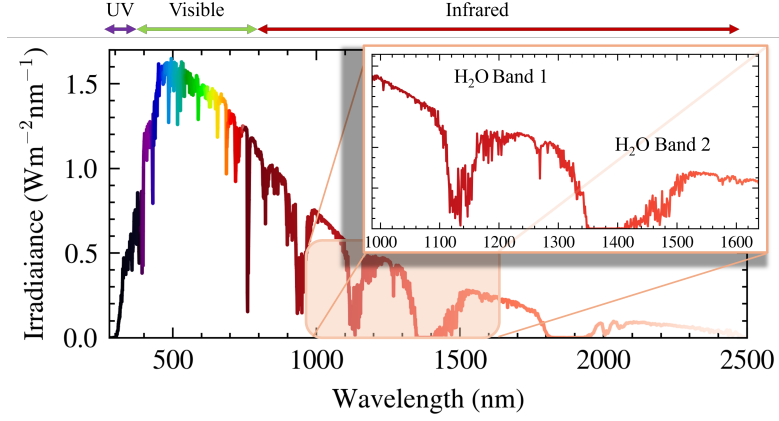


Figure 8: The solar spectrum at ground level, highlighting the two water absorption bands contributing to the spectrum used in this thesis.

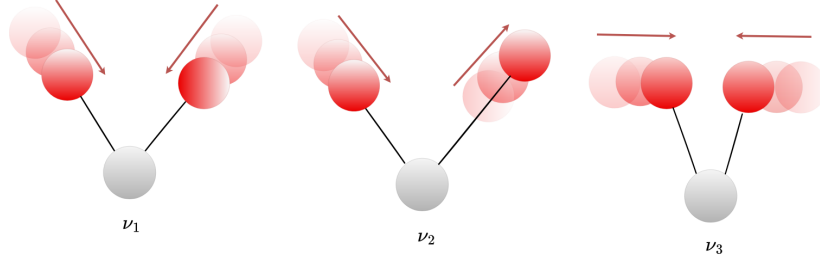


Figure 9: Vibrational modes of water.

water combined with finer rotational bands, allowing the molecule to absorb light and turn it into internal energy [20].

The water molecule has three vibrational modes: $\nu_1 = 2734$ nm, $\nu_2 = 6270$ nm, $\nu_3 = 2662$ nm. Shown in Figure 9 These are the symmetric stretch, bend and anti-symmetric stretch, respectively. Overtones of these yield higher energy transitions, resulting in the bands we find in the short-wave infrared range.

As it turns out, these bands can be utilized in the Lidar to solve for the molecular absorption coefficient for water α_m . Knowing the strength of each absorption line, the absorption coefficient can then be used to determine the concentration of water molecules in the atmosphere. This is done using the DIAL (Differential Absorption Lidar) technique, where two nearby wavelengths, one of which is at a known absorption line (the on-line) and the other one is not (the off-line) [8]. Similar to the slope method, one can use the exponential decay of the signal to determine the absorption coefficient of the on-line wavelength. The lines are close enough such that aerosol influence are assumed to be the same. Comparing the two signals, the difference is then only the attenuation due to molecular absorption. The difference between the extinction coefficients between the off and on-line wavelengths are:

$$\Delta\alpha = N\Delta\sigma \quad (15)$$

where N is the molecular number density and σ is the extinction cross section difference:

$$\Delta\sigma = \sigma(\lambda_{on}) - \sigma(\lambda_{off}). \quad (16)$$

The molecular number density can then be extracted from the Lidar equation as

$$N = \frac{1}{2\Delta\sigma} \left[\frac{d}{dr} \ln \left(\frac{P_{on}}{P_{off}} \right) \right]. \quad (17)$$

3 Methodology

3.1 Setup

The design of the instrument used is described in [2]. A sketch of the setup is shown in Figure 10. The instrument is a bistatic elastic Scheimpflug Hyperspectral Lidar. Bistatic means it has a separate receiver and emitter, Scheimpflug in that it uses the Scheimpflug principle as described before. It is hyperspectral in that it uses a grating to split the wavelengths such that spectral measurements can be made.

3.1.1 Supercontinuum Source

The light source is a diode-pumped solid-state laser with a characteristic Nd:YAG line of 1064 nm, which has been broadened by propagation through a nonlinear fibre. The resulting spectrum is shown in Figure 11. As can be seen, the spectral range is 400 - 2400 nm. The Nd:YAG repetition rate is 80 MHz, the pulse duration is 400 fs, and the max pulse energy is 50 nJ.

The setup is shown in Figure 10 where the beam is expanded with a negative lens and the visible part of the spectrum is stripped using a dichroic mirror and a long-pass filter. The beam is then collimated with a positive lens and emitted at a power of around 3 W.

3.1.2 Receiver

The instrument also contains a receiver which consists of a Newton telescope and a spectral analyser. This analyser projects the incoming light on a 2d InGaAs array. The first dimension is range which is mapped from the angle of the incoming light using the Scheimpflug principle and projecting it on a tilted slit. The second dimension of the detector is for wavelengths separated by a grating in the spectral analyser. The range part has 640 pixels and the wavelength part has 512. The intensity is stored as 14-bit data, and the camera can be adjusted in exposure time, refresh rate, and gain.

The spectral range of the detector is 600 nm (1000 - 1600) nm. The spatial range of the system is theoretically infinite, though at high ranges, range resolution deteriorates.

3.1.3 Line of Sight

The instrument is positioned in the tower lab of Fysicum in Lund, and is directed north-east in a horizontal direction, shown in Figure 12. The light propagates at a safe distance above the ground and terminates at a chimney at Kemicentrum. It also passes over the roof of the Ecology building, where controlled-release measurements can be made. The line of sight is secured with an infrared monitor camera, such that the light from the instrument can be tracked.

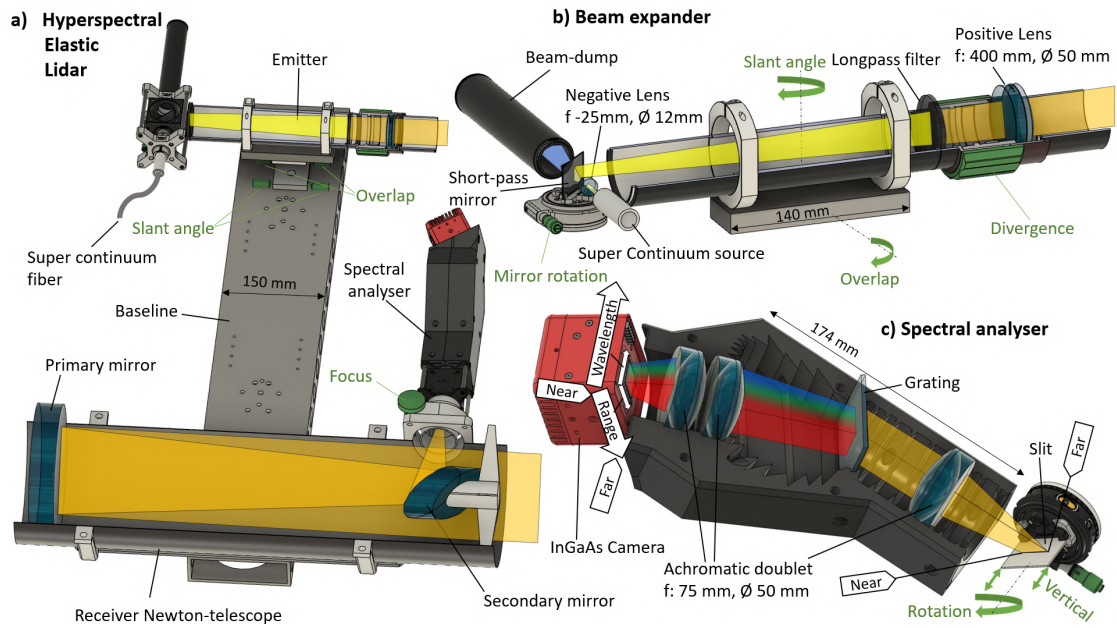


Figure 10: Description of setup, adapted from [2]. The beam expander and receiver telescope are separated by a baseline. Light from the beam expander is stripped of its visible part and emitted. Backscattered light is collected by the Newtonian telescope, projected on a tilted slit, and dispersed onto the 2D focal array.

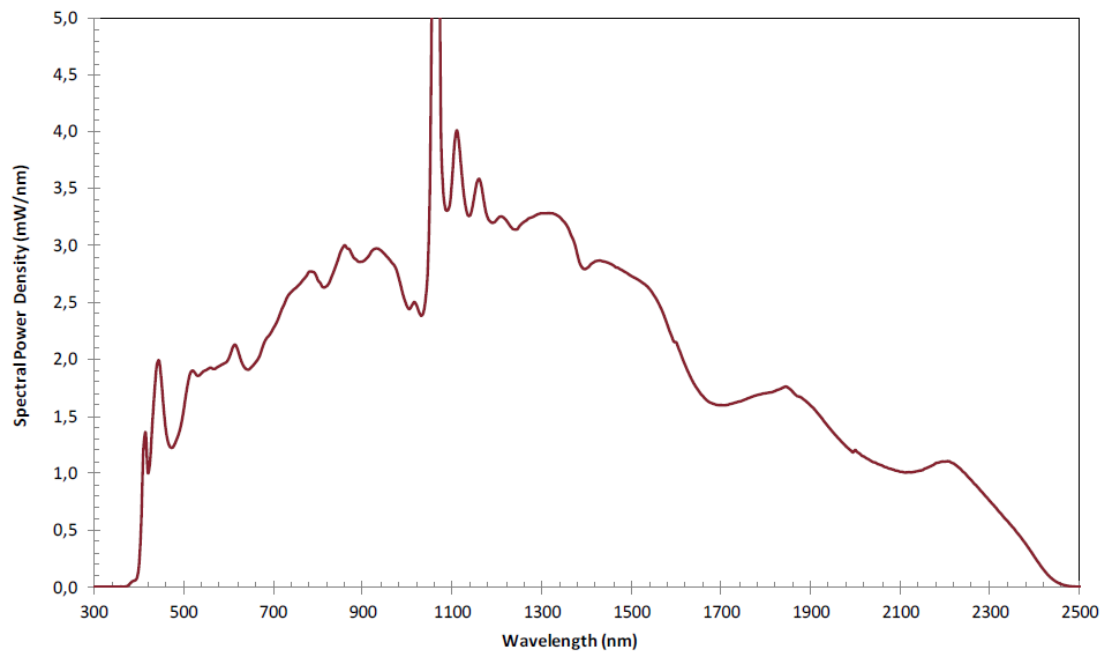


Figure 11: Spectrum produced from the supercontinuum fiber laser SC400 Fianium, (Nordiske Kabel og Tråadfabriker (NKT), Denmark). Figure adapted from datasheet, NKT

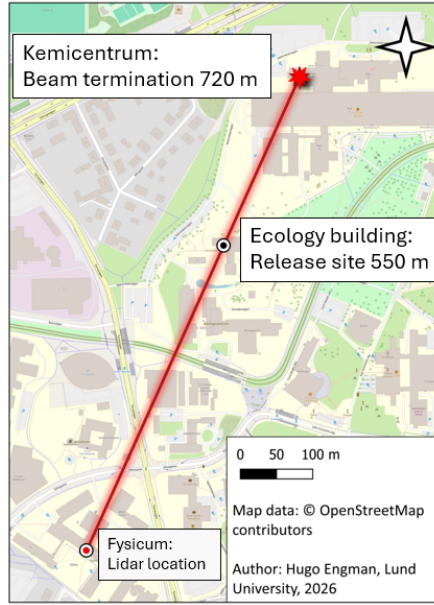


Figure 12: Line of sight described. Lidar beam is emitted from Fysicum, passing over the ecology building where controlled aerosol releases can be conducted before terminating at Kemicentrum.

3.2 Measurement Principle

As described in the previous section, the system operates by emitting supercontinuum light along the line of sight, which is partially backscattered by atmospheric particles and collected by the receiver. The light is then collected and dispersed onto the 2d detector, where each row corresponds to a certain range along the beam path and each column to a specific wavelength.

The detector pixels thus measure the backscattered light at each wavelength and distance. Backscatter and extinction cause variations in signal intensity over the range. Spectral variations occur due to wavelength-dependent effects, such as Mie scattering or molecular absorption bands.

The termination measures the beam intensity at the end of the line-of-sight and serves as a reference for integrated attenuation along the beam path. Extinction caused by particles along the beam path will reduce the termination signal accordingly. Thus, the retrieval of range and spectrally resolved backscatter and extinction is possible in a single acquisition.

The hyperspectral acquisitions are then mapped over time to generate a three-dimensional cube that now contains spatial, spectral, and temporal information about the atmospheric return.

3.3 Data Acquisition

The hyperspectral data cubes were recorded in LabVIEW [21] through a script where instrument parameters could be adjusted such as exposure time, gain, number of exposures and refresh rate. Preprocessed data could be visualised, allowing parameters to be optimised to balance spectral and temporal resolution as well as SNR.

To avoid saturating the termination signal, gain and exposure time were required to be sufficiently low. In general, gain was kept low while the exposure time was increased to stabilize SNR. Exposure time could be increased up to 500 ms, but was typically kept at 200 ms as it allowed for good temporal resolution.

Measurements were made by sequentially stacking exposures in raw data files uploaded to a computer for storage and processing. Between files, a dead time occurred while the file was written to disk. For a 200 ms exposure time file containing 200 images, resulting in a 40 s measurement time, the dead time was around 24 seconds.

Inherent in the detector was dark current which added a base-level to the signal, increasing over time as the detector heated up, and then plateaued. Within each acquisition, the dark current was found to be the highest in the beginning, immediately after transferring the previous acquisition to the computer. The contribution varied spatially over the detector and was particularly elevated along the edges due to non-uniform heating. Dark current measurements were made by regularly blocking the emitter and recording the background detector signal, which was then subtracted in pre-processing.

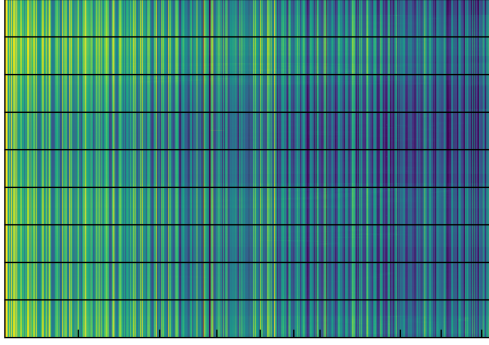
Another noise source was solar background radiation as parts of the solar spectrum overlaps with that of the instrument, see Fig 8. The measurements were thus only conducted after sunset. However, solar background contributions were still observed up to around an hour after sunset.

3.4 Pre-processing and Calibration

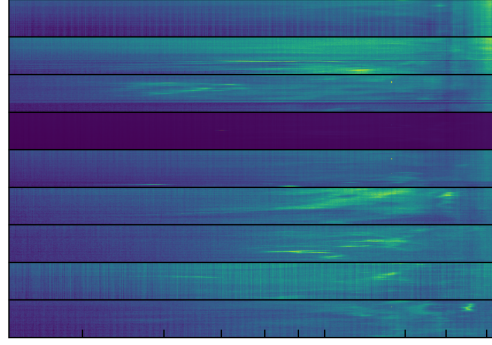
Due to detector noise and background radiation, data preprocessing was necessary, especially for low signal strengths.

The recorded files were first stored in three-dimensional arrays, allowing each data dimension to be treated individually. Due to dark current drift, particularly between files, dark current was subtracted from each exposure of each file with a background measurement from the same file. Further, the dark current was subtracted to obtain a similar signal baseline for each file. Sequential files were then combined such that the entire time evolution of the measurement could be treated.

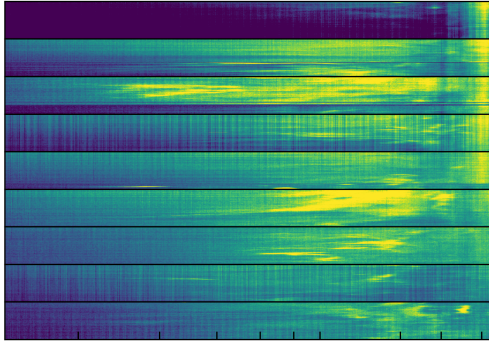
To improve photon statistics, primarily temporal averaging over multiple files was applied but also spatial and spectral averaging was done. For faint signals, rescaling and image filtering were applied to improve visualisation. Different steps to enhance aerosol signals are shown in Figure 13.



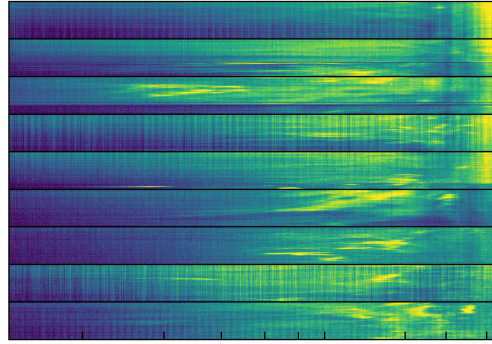
(a) Raw data.



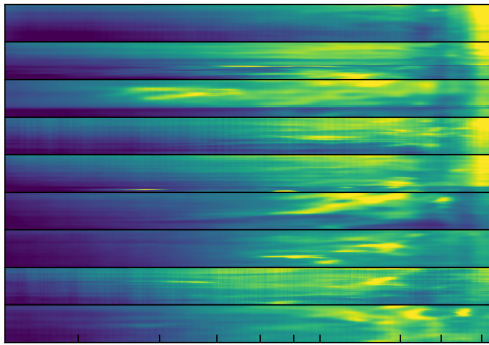
(b) Background subtraction of dark current.



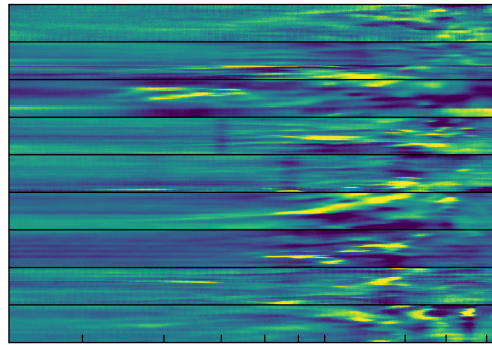
(c) Subtracting the baseline signal level to bring the baseline to 0.



(d) Normalise the signal by its 99th percentile with respect to noisy highs.



(e) Apply bilateral filter to keep sharp edges while reducing noise.



(f) Subtract the mean of each row and column beforehand to remove stripy noise.

Figure 13: Different re-scaling and filtering techniques to enhance time-range visualization

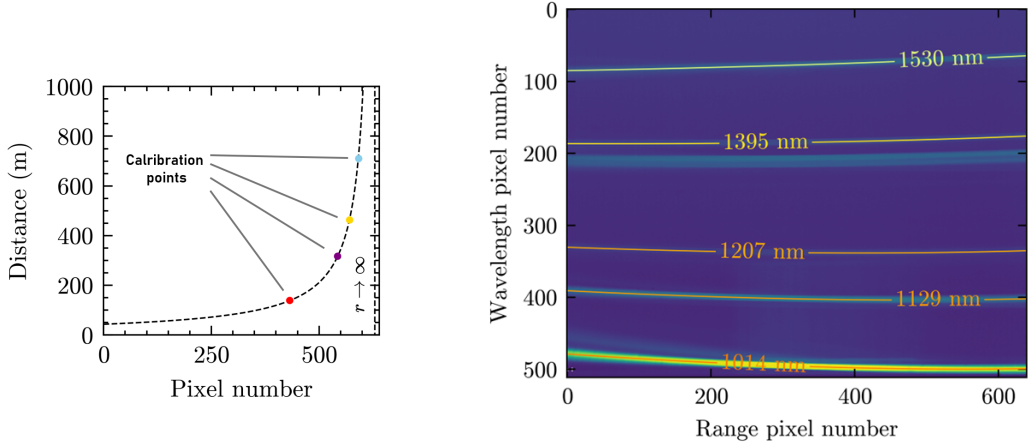


Figure 14: Peaks of Hg used to calibrate wavelengths along the 2d chip (Left). Distance calibration using four calibration points at different distances.

Calibration was required to map each pixel on the 2d detector to its corresponding wavelength and range. Spectral calibration was done with a Hg lamp, which produced spectral lines throughout the entire pixel range in figure 14. These lines are curved due to aberrations in the optics. Along the wavelength axis, the bending occurs due to the smile effect and on the range axis, the bending is due to the keystone effect [2]. To find the right form for this curve, multivariate polynomial curves were fitted to each line using a least squares method. The number of terms used was added until no significant improvement on the fit was obtained. The resulting wavelength calibration had the form:

$$\lambda = \lambda_0 + \lambda_1 p_\lambda + \lambda_2 p_r + \lambda_3 p_\lambda p_r + \lambda_4 p_\lambda^2 + \lambda_5 p_r^2 + \lambda_6 p_\lambda p_r^2 + \lambda_7 + \lambda_8 p_\lambda^3 + \lambda_9 p_r^3 \quad (18)$$

The distance scaling of the Scheimpflug lidar is based on triangulation and has the functional form of an arctan function [2]:

$$r = \frac{b_l}{\tan(\Phi_{slant} + \theta_{FOV} p_r + \psi_{shift} p_\lambda + \psi_{fan} p_\lambda p_r)}, \quad (19)$$

where p_λ, p_r are normalized pixel positions. Φ_{slant} is the slant angle and θ_{FOV} is the field of view of the instrument. b_l is the baseline distance and ψ_{shift} and ψ_{fan} are parameters accounting for offsets given by artifacts. For the calibration I used the same b_l , ψ_{shift} and ψ_{fan} parameters as in the previous work [2]. But the ψ_{fov} and Φ_{slant} parameters were calibrated by illuminating targets at different ranges throughout the beam path where the distances were measured in Google Earth. Again, the parameters were determined using a least squares algorithm. The fitting is shown in Figure 19.

3.5 Experimental measurements

To investigate different scattering and absorption processes, measurements were carried out for a range of atmospheric targets, including ambient aerosols, controlled aerosol plumes, fog, precipitation and molecular absorption. Each measurement was made to obtain wavelength-dependent properties of different targets to examine the applicability of the spectral range of the instrument.

3.5.1 Hydrometers

Measurements of fog, snow and rain were made to investigate high extinction and larger atmospheric targets. Fog measurements were carried out to examine atmospheric conditions with strong attenuation and high optical depth. This allowed for a better understanding of spectrally dependent scattering where single-scattering assumptions are no longer valid. By tracking the fog clouds' movements along the beam path through time, wind velocity could be determined as well.

Falling snow was also measured to determine the scatter from larger non-spherical scattering targets which deviates strongly from Mie assumptions. To assess the extent of the dispersive effects, their colours were examined. By reducing the detector's exposure time, individual snowflakes could be captured and their colour determined. The snowflakes were then binned to their respective color range and each of their extinction coefficients was measured.

Rain measurements were also performed to examine scattering of isolated water droplets and compare the signal to that of fog and snow.

3.5.2 Ambient measurements

Ambient atmosphere without precipitation was also examined under different weather conditions to measure which had the largest effect on the hyperspectral signal. For each measurement, variables such as wind speed and direction, temperature, relative humidity and aerosol concentrations were noted.

PM10 (particulate matter under 10 microns) concentrations were used to measure the impact of ambient aerosols on the measured data. Specific days with high aerosol loading were chosen to fully quantify these effects. Relative humidity and temperature were also monitored to determine water vapour concentration as a reference for DIAL measurements.

3.5.3 Plumes

Controlled aerosol plumes were released into the beam and measured to obtain atmospheric returns from known scattering targets under repeatable conditions. The plume release site was the Ecology building as shown in the Line-of-sight Figure 12, at a range of around 550 meters from the instrument in its path.



Figure 15: Multiple plumes emitted into the beam, producing scattering.

The experiment included participants who were present by the Lidar for operation and monitoring, and three controlled the plume releases by the release site. Each acquisition sequence was designed to contain a clear termination signal, a dark background measurement and a plume signal within the same file. The plume material, as well as the local time was recorded for each plume.

The obscured termination signal was used as a reference to measure attenuation during the plume release to obtain the optical depth. A neoprene target with a reflectance of around 2% was used to calibrate the backscatter signal. To account for varying atmospheric conditions, this calibration was done multiple times throughout the experiment. To generate aerosol plumes, a compressor and nozzle were used to create clouds of dust and sprays into the beam. The plume release can be seen in figure 15.

3.6 Signal Analysis

3.6.1 Extinction

To determine extinction, the two-way transmission to the termination was measured at a clear signal and an obstructed signal. Comparing the lidar equation 8 between the two scenarios at the termination

$$P_{clear}(r_{term}) = P_0 C \frac{\beta_{clear}(r_{term})}{r_{term}^2} \Delta r_{term} \exp\left(-2 \int_0^{r_{term}} \alpha_{clear}(r) dr\right), \quad (20)$$

and

$$P_{blocked}(r_{term}) = P_0 C \frac{\beta_{blocked}(r_{term})}{r_{term}^2} \Delta r_{term} \exp\left(-2 \int_0^{r_{term}} \alpha_{blocked}(r) dr\right), \quad (21)$$

it can be seen that $\beta_{clear}(r_{term}) = \beta_{blocked}(r_{term})$ as no plumes are released at r_{term} . Dividing the two equations then yields

$$\begin{aligned} \frac{P_{clear}(r_{term})}{P_{blocked}(r_{term})} &= \exp\left(-2 \int_0^{r_{term}} [\alpha_{clear}(r) - \alpha_{blocked}(r)]\right) = \\ \exp\left(-2 \int_0^{r_{term}} [\alpha_{mol}(r) - (\alpha_{mol}(r) + \alpha_{aer}(r))]\right) &= \exp\left(2 \int_0^{r_{term}} \alpha_{aer}(r)\right). \end{aligned} \quad (22)$$

Solving for the integrated extinction we get,

$$\frac{1}{2} \log\left(\frac{P_{clear}(r_{term})}{P_{blocked}(r_{term})}\right) = \int_0^{r_{term}} \alpha_{aer}(r) = AOD, \quad (23)$$

where AOD is the aerosol contribution to optical depth.

3.7 Backscatter

To determine the backscatter, the lidar equation was used again but at $r = r_{release}$ for the release site. Now, the signal of the plume was compared to the reflected signal from the neoprene board with known diffuse reflectance of 2% to obtain a calibrated plume signal. The plume was assumed to be spatially flat, hence only β_{plume} and $\beta_{neoprene}$ are different between the two signals. Thus,

$$\begin{aligned} \frac{P_{plume}(r_{release})}{P_{neoprene}(r_{release})} &= \frac{\beta_{aer}\pi}{R_{neoprene}} = \frac{\beta_{aer}\pi}{0.02} \\ \implies \beta_{aer} &= \frac{0.02P_{plume}(r_{release})}{\pi \cdot P_{neoprene}(r_{release})} \end{aligned} \quad (24)$$

3.8 Lidar ratio

Using AOD and the integrated backscatter coefficient β , the averaged Lidar ratio could then be determined

$$LR = \frac{AOD}{\beta}. \quad (25)$$

3.9 DIAL

For DIAL measurements, averaging over multiple lines was done as the spectral width of water absorption lines is narrower than the resolution of the detector. The atmospheric water vapour concentration was determined from relative humidity and temperature to use as a reference for the retrieval. Due to low signal strength,

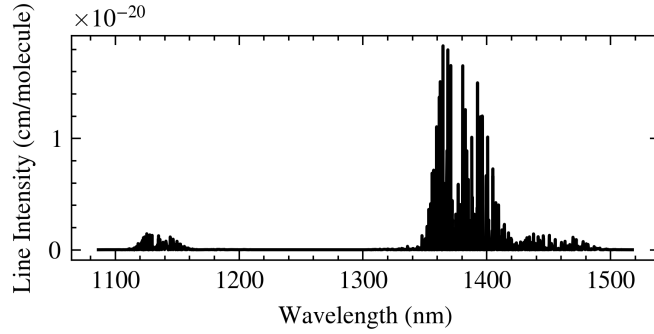


Figure 16: Line intensities of absorption bands of water in the short-wave infrared, showing two distinct wavelength regions of high absorption.

temporal averaging was done over the whole measurement period. To obtain the off and on-line pixel positions, the calibration method was used to obtain the lines of constant wavelength across the detector, similar to those of Hg in 14 and interpolation was done between pixels to obtain the precise line. A Gaussian filter was then applied to smooth the signal. At first, the lines were chosen to occur at a single wavelength to optimise spectral precision, but this resulted in noisy signals, so averaging over wavelength was performed.

The stronger absorption band (1350-1370 nm) and the lower one (1100-1160 nm) were then chosen as on-bands for multiple DIAL measurements, as well as an off-band in between the two (1200-1300nm), where absorption is low. An additional off-band (1000-1100 nm) was used to measure the relative attenuation between the two off-bands and to assess other effects besides absorption. The dial equation was then used in combination with an averaged absorption cross-section from spectroscopic data in Hitran together with the MT_CKD water supercontinuum model[20], the overtone cross sections in the spectral range were calculated, shown in Figure 16 to determine the number concentration of water vapour which was then converted to mass concentration to compare it to the averaged water concentration noted for the day.

3.10 Simulations

To further investigate the potential benefits of hyperspectral lidar, a simulation was done to observe the improvement of retrieval of microphysical properties for three wavelength information from typical Nd:YAG harmonics compared to 64-band hyperspectral (1000-1600 nm) data similar to that of the instrument. An additional 64-band spectrum was investigated between (300-1000) nm to compare hyperspectral retrieval between different spectral ranges.

To simulate data, artificial backscatter and extinction coefficients were calculated for two different particle distributions of sea spray in two different size modes, retrieved from OPAC's aerosol model [22]. The log-normal parameters of these are $r_{mod}, \sigma = 0.209, 2.03$ and $1.75, 2.03$ microns respectively. From these, the backscatter and extinction coefficients were determined through a Mie scattering

model [23].

The specific particle distributions were chosen to obtain size parameters ($r/\lambda \approx 1$) to probe the Mie scattering regime. Both distributions were chosen to have similar backscatter and extinction coefficients by adjusting number concentrations. In the retrieval algorithm, the number concentration was not estimated to keep the number of free variables low.

Synthetic data was then produced by adding 20% Gaussian noise to α, β to produce data similar to that retrieved by Lidar. By increasing the number of sampling wavelengths from 3 to 64, a corresponding noise factor was multiplied which was $\sqrt{64/3}$ to account for lower photon statistics per pixel, as the total signal is distributed among more wavelengths.

To retrieve the mode and width of the distributions, different parameter values were sampled and used in Mie calculations to determine α and β . The log likelihood was then evaluated with respect to the true α, β using eq. (2.6.1). This was implemented using a Monte Carlo Markov Chain algorithm in Python, where parameters are sampled by "walkers" that take random steps in parameter space. The steps are either accepted if the posterior is higher than the previous step or otherwise rejected. By retrieving the sample chains of the walkers, a distribution was generated of the most likely parameter values.

The median of these was then determined to serve as the most likely parameters. These were compared to the original parameters to measure the effectiveness of the inversion. A more quantitative approach was used, in which the cumulative particle size distribution was determined and a Kolmogorov-Smirnov test was performed. This is a goodness-of-fit test, which serves as a quantitative measure of the difference between retrievals.

The algorithm is initially applied to each set of wavelengths for unimodal distributions, then to bimodal distributions. This was done to investigate the benefits of microphysical retrieval when aerosol mixing is introduced, resulting in more complicated spectral dependence of the distribution.

4 Results

4.1 Hydrometeors

The strong scattering strengths of hydrometeors proved a good probe for hyperspectral measurements. Different types of precipitation yielded greatly different scattering effects, shown in Figure 17. Fog produces large, diffuse clouds with high signal strengths, while snowfall produces small, concentrated backscatter profiles. Rain shows the lowest backscatter signal but reveals sharp features across the range, blurred out by extinction from raindrops.

4.1.1 Fog Clouds

Illuminated fog produced strong broadband backscatter and attenuation. The resulting data are shown in Figure 18, which highlights the variation in the spectral, spatial, and temporal behaviour of different fog clouds.

A single time-frame (a,b) shows spectral signatures which vary over the beam path, these represent wavelength-dependent backscatter and attenuation from multiple clouds. A decrease in intensity over the range, followed by a weak termination signal, highlights strong attenuation and high optical depth.

The spectral signatures are most prominent in the pump signal at 1064 nm and in the main water absorption band around 1400 nm. The beam is strongly attenuated in this wavelength regime, resulting in the spectrum produced by the termination (e) becoming flat.

Over time, clouds vary along the beam path (c). They move to higher range values as time increases, indicating their wind speed and direction. The signal of clouds and termination at the beginning of the measurement (e), appear correlated, increasing and decreasing together. This occurs up to around 25 seconds where cloud signal drastically increases and termination decreases, showing anti-correlation instead.

Due to strong presence of water in the atmosphere in this measurement, molecular attenuation can be seen to dominate spectrally at water vapour absorption bands. This makes it impossible to retrieve spectrally resolved signals at the places where it is extinguished completely, reducing the spectral capacity of retrieval. The otherwise strong spectral signatures throughout the range, however, make fog a good candidate for hyperspectral detection. The correlation between clouds and termination indicated that some of the cloud scattering is detected as termination return, which could be a sign of multiple scattering.

Furthermore, wind speed was determined for the fog, shown in Figure 19, which shows speeds between 1-3 m/s. For atmospheric studies, this could be used to determine the origin of the fog.

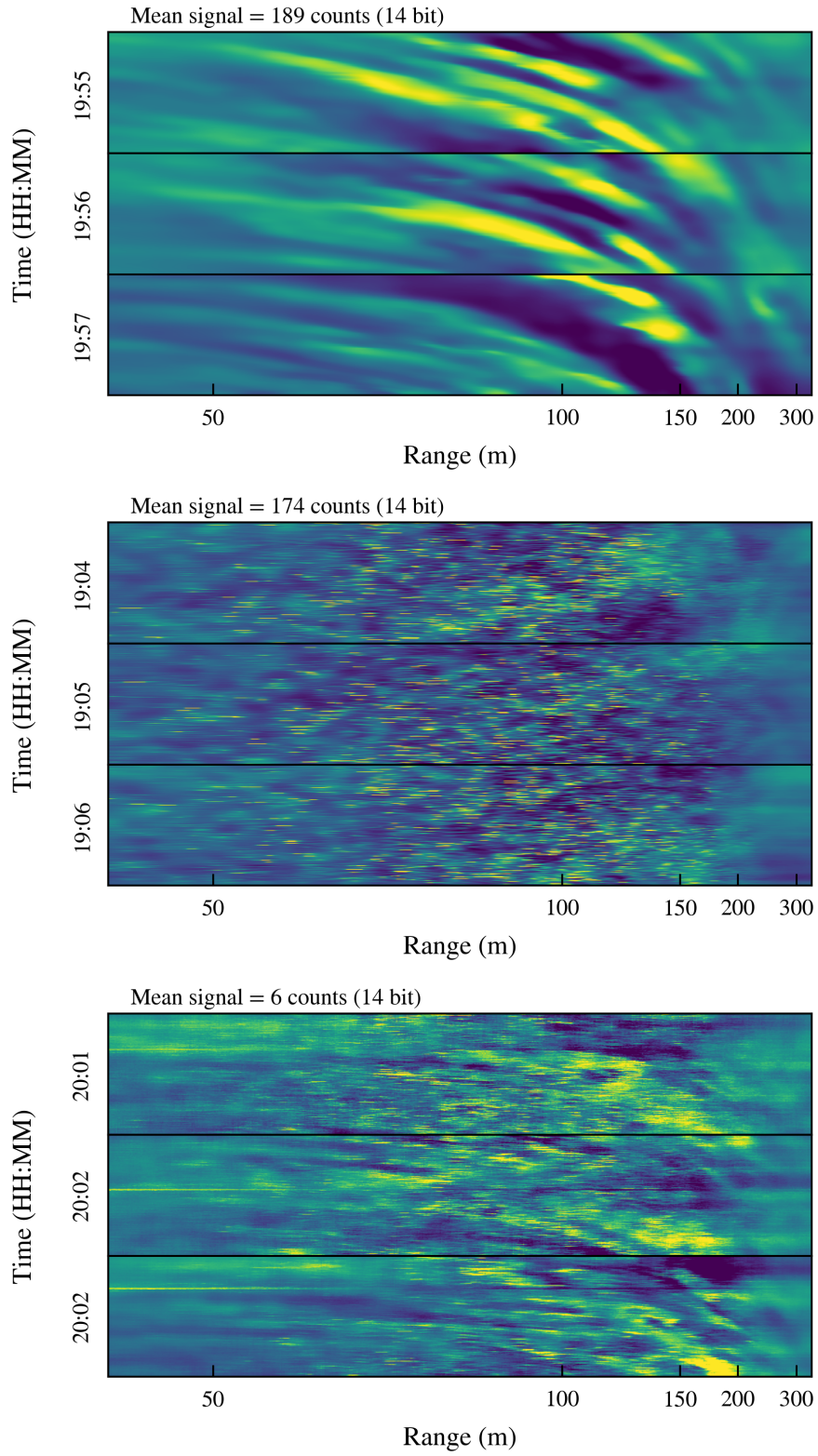


Figure 17: Monospectral time-range map of (Top) fog, (middle) snow, (bottom) rain.

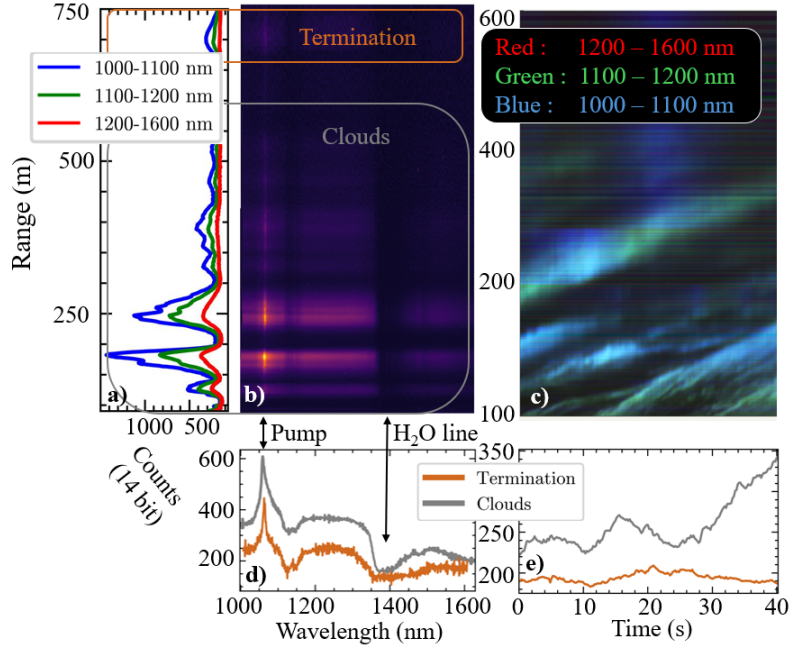


Figure 18: A single snapshot exposure of the elastic lidar. a) Intensity of different bands over a selected range. b) Heatmap of backscattered light intensity on the focal plane array. c) Full waveform echoes of selected spectral bands. d) Continuous spectrum of natural clouds and the termination echo. e) Clouds and termination signal over time.

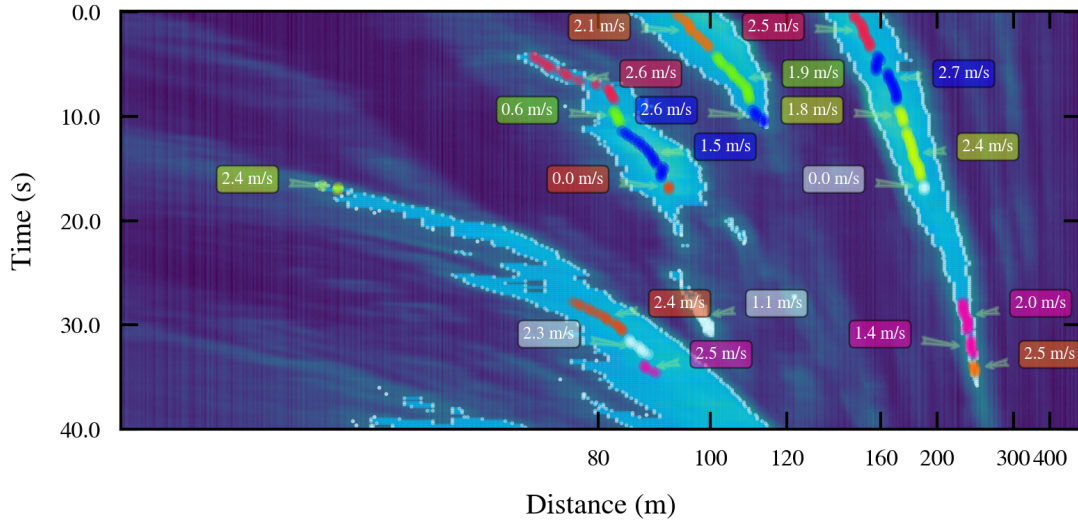


Figure 19: Wind speed determined for different detected fog clouds along the beam path.

4.1.2 Snow

Snow measurements produced strong spectrally varying scattering from isolated ice crystals. Nonspherical scattering produced significantly sharper wavelength dependencies compared to the fog and aerosol spectra as well as high signal strengths.

Reducing the exposure time of the detector revealed individual snowflakes, each spectrally resolved, shown in Figure 20. Each snowflake produced a unique spectrum which could be binned in different colour bands. Three of those spectra are highlighted in Figure 21. Each of them produces a high-intensity, uniquely different spectral signature. All three snowflakes showed strong attenuation due to water absorption, while one stands out because it lacks a distinct pump signal.

These strong, wavelength-dependent spectral curves indicate a new scattering regime absent in the fog measurement. The high backscatter intensity for such a low exposure time indicates a highly reflective surface, as is the case with flat surfaces that produce specular reflection.

As snowflakes are non-spherical, the strong wavelength dependence signifies a dispersive effect at the interface. Snowflakes typically produce dispersion by refraction, which cannot be purely the case here, as the light is not scattered forward but backwards. Thus, iridescent snowflakes may be produced by a combination of refractive and specular reflective effects from multiple scattering. In any case, strong refractive or specular reflective effects from flat surfaces must be strongly correlated with snowflake orientation.

Using the slope method and binning three bands, the extinction coefficients of these bands were determined in Figure 22. The red, green and blue bands were adjusted slightly such that the red was the broadest of these and thus had the highest counts. The extinction coefficients showed 1.9 km^{-1} for the green and blue bands while 1.1 km^{-1} for the red.

Strongly varying spectral features for individual ice crystals not present in aerosol and fog data demonstrate the additional information that can be obtained through hyperspectral lidar measurements.

4.2 Plumes

Controlled aerosol plumes provided known, high backscatter targets under repeatable conditions which illuminated the entire spectrum. Unobscured termination signals together with a known reflective neoprene target, enabled retrieval of backscatter and extinction coefficients.

One file of hyperspectral plume measurements is shown in Figure 23, where five plumes can be identified in local ranges of high backscatter as well as the attenuated termination at 700-800 m. A decrease in the termination signal follows from an increased plume signal resulting from increased optical depth in the aerosol clouds. The termination signal showed broadening, indicating reduced spatial resolution at large distances. An oscillation in space of both termination and plumes is

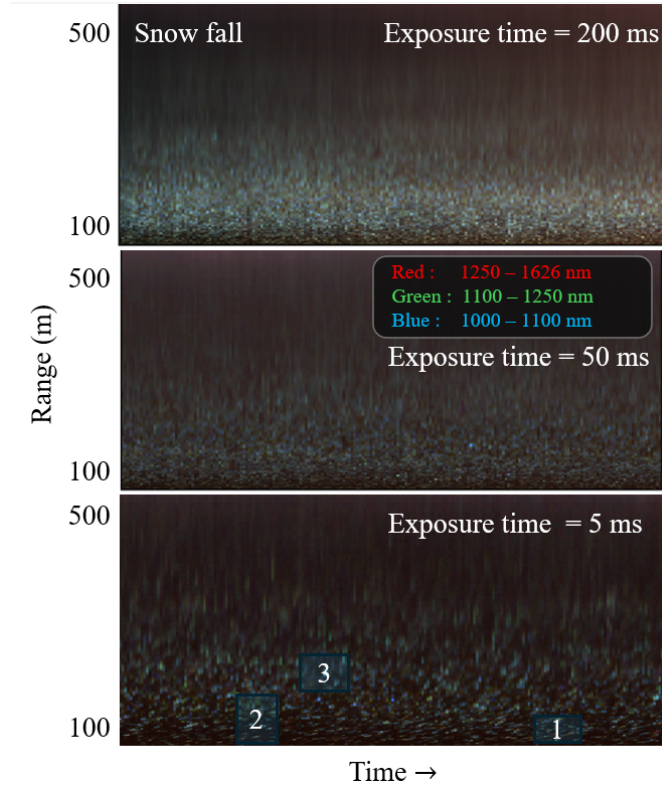


Figure 20: Snow Flakes with different exposure times highlighting the tradeoff between signal strength and temporal resolution

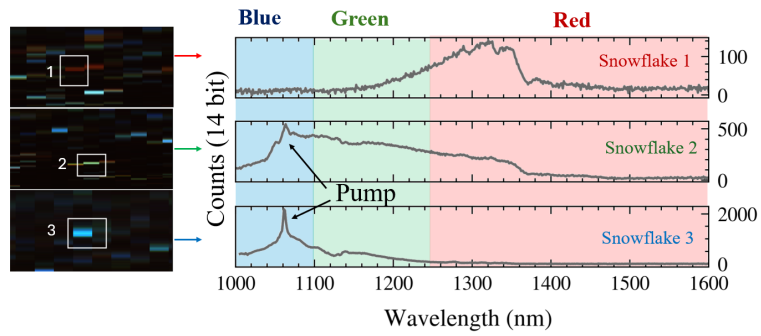


Figure 21: Hyperspectral lidar observation of shimmering snowflakes. Each ice crystal produces a different spectrum.

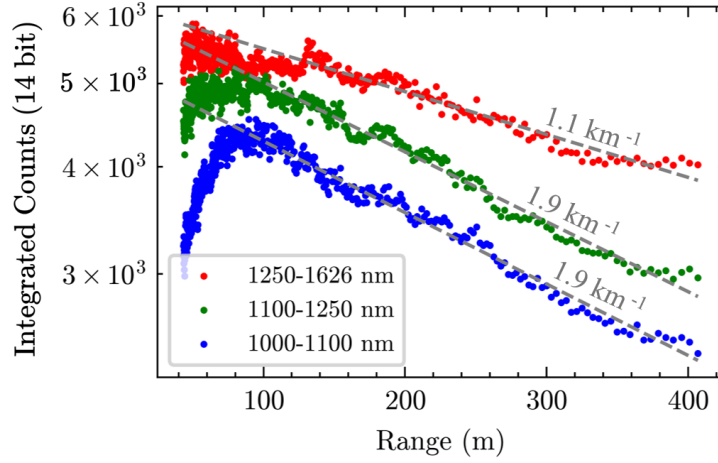


Figure 22: Summed intensity and fitted extinction coefficients from snowflakes of three different spectral bands.

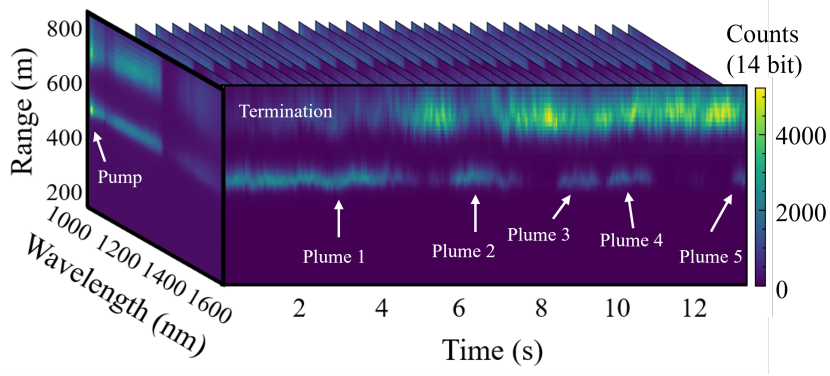


Figure 23: Sea spray data cube highlighting the capabilities of the system in three dimensions: range, wavelength, time and intensity

attributed to turbulent air near the ground, which changes the refractive index. Despite a long measurement range, strong signals were obtained, around 3000 counts for the plume, and the entire spectral range was covered except for the water absorption bands.

A close-up of the generated water plumes is shown in Figure 24. Using the unobstructed terminal signal as a reference, the beam transmission through the plume to the termination could be measured over time. Using the neoprene to calibrate the signal strength, the integrated backscatter was determined over time. The transmission is spectrally even while the backscatter showed reduced intensity at higher wavelengths, corresponding to the red spectral band.

Each plume was measured in its backscatter and extinction coefficient over time to determine the Lidar ratio. The water plume mentioned previously is shown in Figure 25. Outside the water absorption lines, the Lidar ratio shows a relatively continuous behaviour, ranging from 30 to 50 sr. The smooth spectral behaviour

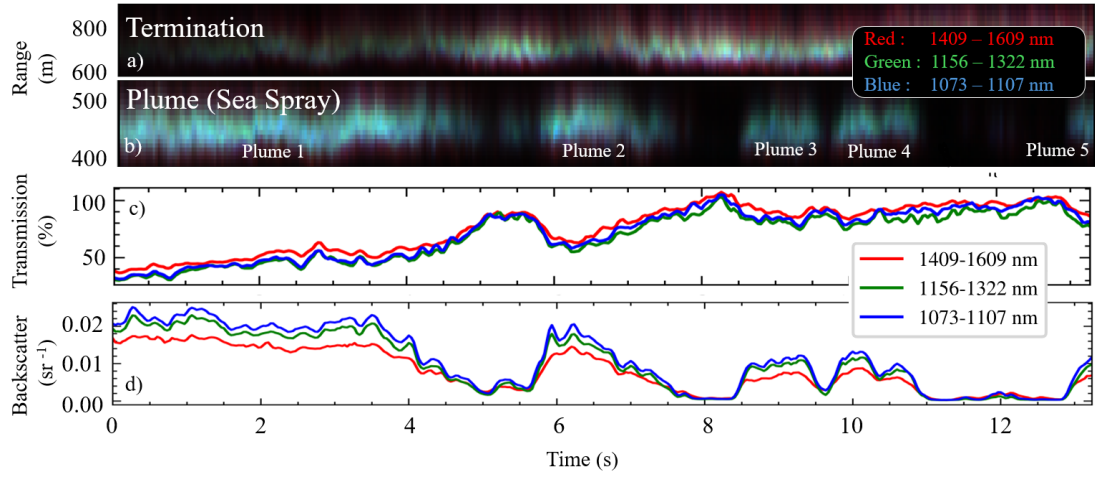


Figure 24: a) False colour composite time-range maps of a) the termination and b) plume of sea spray, highlighting individual plumes in three different spectral ranges. From these spectral bands: c) one-way transmission to termination and d) backscatter from plumes.

indicates diffuse scattering from a distribution of scattering particles.

To better highlight features in the Lidar ratio, the sea spray was measured in its backscatter and Lidar ratio with respect to Aerosol Optical Depth in Figure 27. The backscatter coefficient increased monotonically with the aerosol optical depth as expected, corresponding with a Lidar ratio of 40-50 sr. At larger optical depths backscatter flattened and Lidar ratio increased significantly. This deviation from linear behaviour suggests multiple scattering effects at large aerosol concentrations.

Lidar ratios of plumes of different materials are compared in Figure 26, where Lidar ratios vary between 20 and 50 nm. The lowest is exhibited by Lighter fluid, while the highest is from concrete, with concrete showing higher absorption properties compared to lighter fluid, which shows higher scattering. Each plume's backscatter and AOD vary throughout the spectral range. Especially at wavelengths 1300-1450 nm, where no absorption is present, the variations observed across different materials suggest wavelength-dependent scattering features.

Plumes of different materials are measured against each other more closely in Figure 28. Different spread and clustering of backscatter were observed while all plumes exhibited typical increased backscatter with AOD, reflecting their Lidar Ratios. The largest outlier is concrete dust, showing a constant backscatter of AOD.

Overall, the measurements of controlled plumes demonstrated the hyperspectral Lidar's capability to simultaneously measure extinction and backscatter from concentrated aerosol targets at long distances, while preserving spectral information.

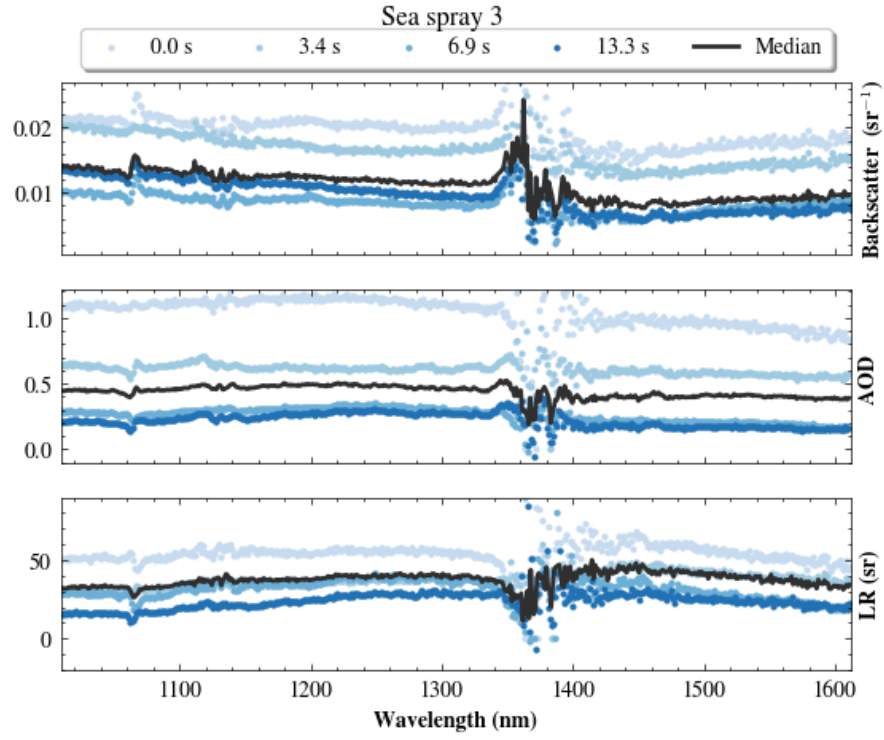


Figure 25: Wavelength-dependent backscatter and extinction coefficient, as well as Lidar ratio of water plume number three, observed at different times.

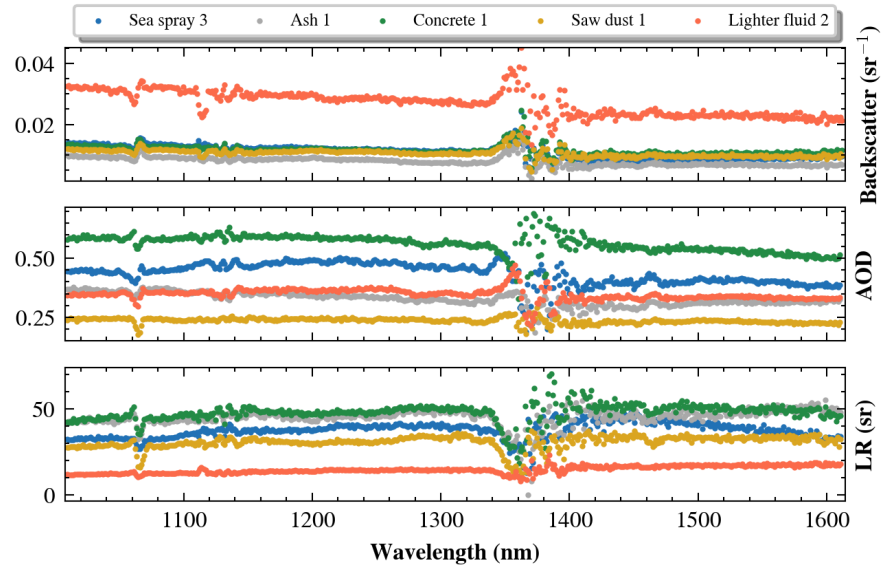


Figure 26: Scattering, extinction and lidar ratio of different generated plumes.

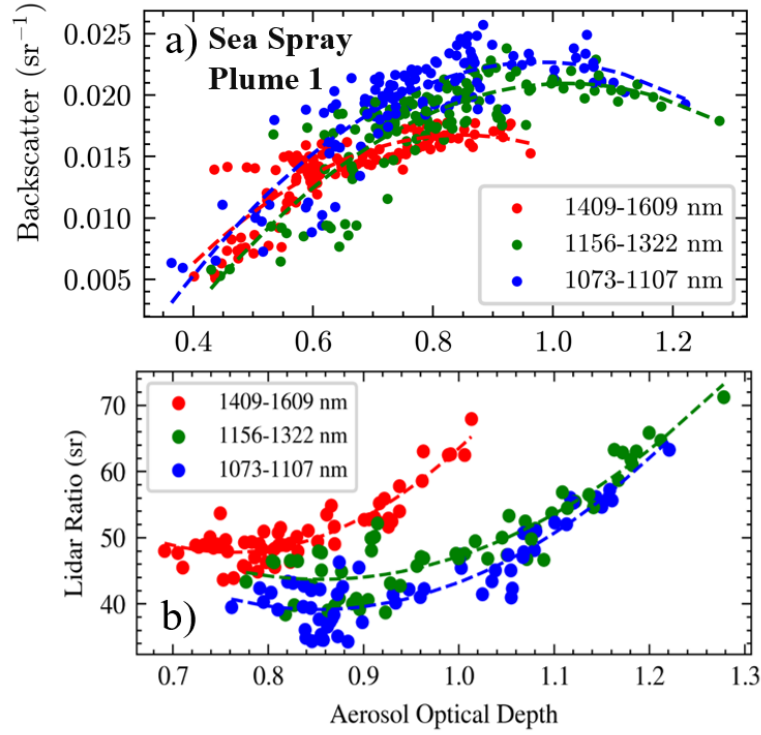


Figure 27: Integrated backscatter of three different spectral ranges for sea spray line fitted with a second-degree polynomial (a) and corresponding lidar ratio and a corresponding second-degree polynomial fit (b).

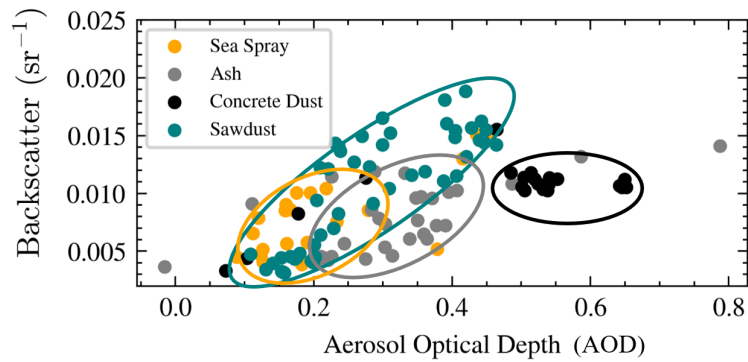


Figure 28: Different materials backscatter against aerosol optical depth, where different materials are circled for clarity.

4.3 Ambient aerosols

For ambient aerosol, low signal strengths were measured, which reduced the possibility of retrieving spectral information due to low photon statistics across the detector. The weather conditions for ambient measurements are shown in Table 1.

Table 1: Atmospheric conditions during hyperspectral lidar measurements.

Date	PM10 ($\mu\text{g}/\text{m}^3$)	RH (%)	Wind (m/s)	Temp ($^{\circ}\text{C}$)	Time
19 Jan	35	90	3 $\nwarrow$	0.5	16:29–17:07
2 Feb	20	60	9 $\leftarrow$	-5.0	17:04–17:45
11 Feb	13	92	7 $\leftarrow$	0.9	17:03–20:19
25 Feb	2	89	4 $\nwarrow$	1.3	17:36–20:33
9 Mar	49	70	3 $\nwarrow$	6.0	18:29–19:07
10 Mar	100	92	1 $\nearrow$	4.0	18:39–19:34
24 Mar	47	100	6 $\nearrow$	9.0	19:48–20:20
25 Mar	10	78	7 $\rightarrow$	5.0	20:30–21:00

Although signal strengths were low aerosol features could be retrieved on February 2, in Figure 29 (top). This was likely due to local dust clouds, swept by car traffic during dry conditions. Similar features were detected on February 25, shown in Figure 29 (middle), where signal strengths were lower and cloud features more concentrated. As this time point occurs later in the day, this suggests less car traffic is a contributor. Another contributing factor is also wind, blowing dust into the beam path. The wind direction was strong westwards on the first day while eastwards on the second.

The highest aerosol PM10 concentrations were found on March 10 and are shown in Figure 29 (bottom), where distinct aerosol clouds could not be observed. Signal strength increases as aerosol concentration increases. A feature of these plots, is that there appears a blurriness over the road, indicating some turbulent air present. The cause of no distinct features despite high aerosol load is likely due to long transport aerosols being well mixed in the atmosphere.

4.4 Water Vapour and DIAL

DIAL measurements provided additional opportunities to retrieve atmospheric spectral information. Broadband features could be used to estimate water concentrations across multiple days.

To obtain a reference for the attenuation across different days, the termination signal could be used to determine relative differences in molecular absorption between measurements. To highlight these differences, the ratio of the termination signal of different days with respect to that from February 25 was plotted, shown in Figure 30. Days with higher water vapour concentrations showed lower termination signals in the absorption bands and days with lower water concentrations had

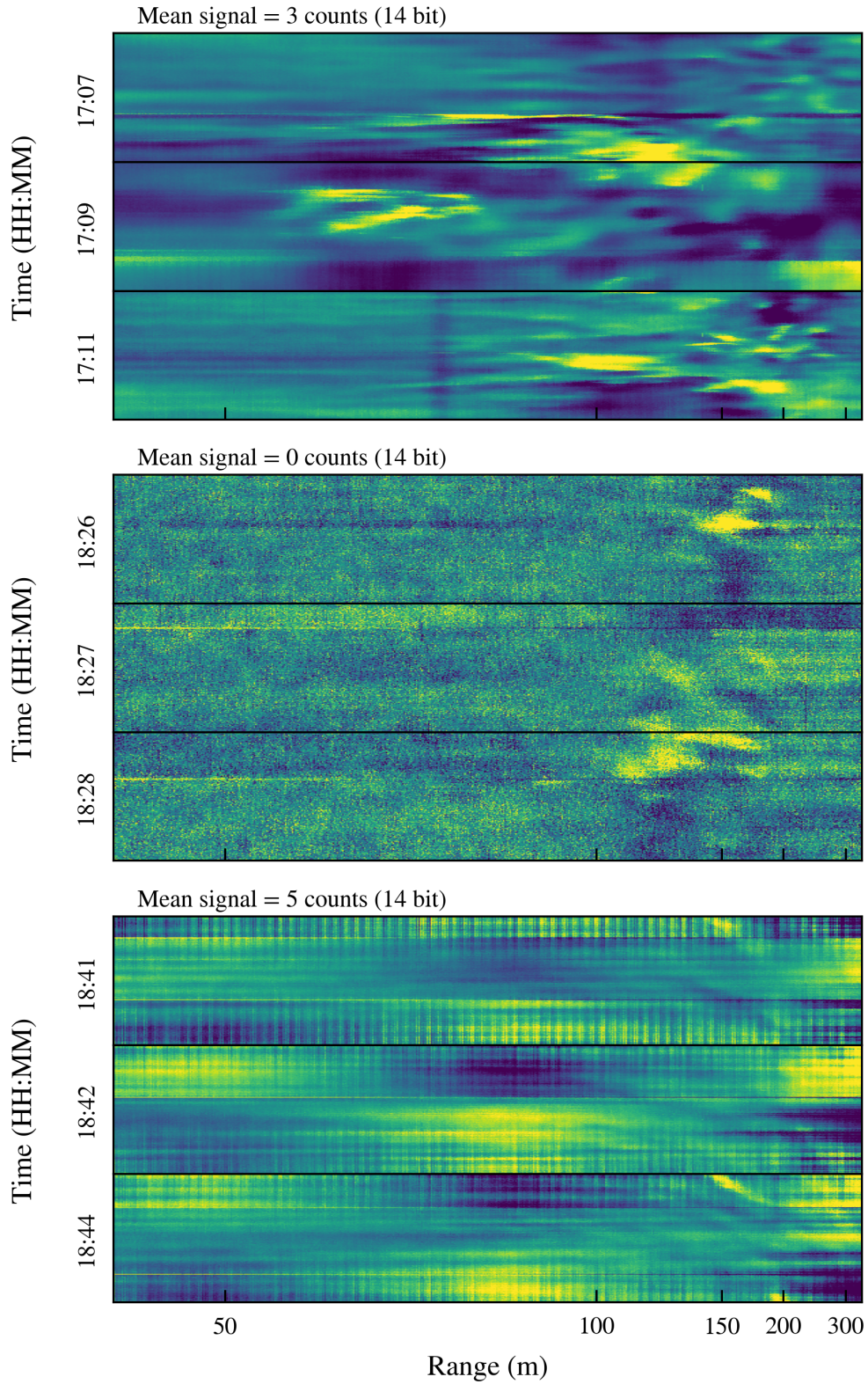


Figure 29: Monospectral image of aerosol loading on February 2 (top), showing cloud-like features. Aerosol loading on February 25 (middle), showing reduced signal strength and noise, with some features at around 150 m. March 10 (bottom), with higher aerosol loading leading to increased mean signal also blurred features at 100-150 meters.

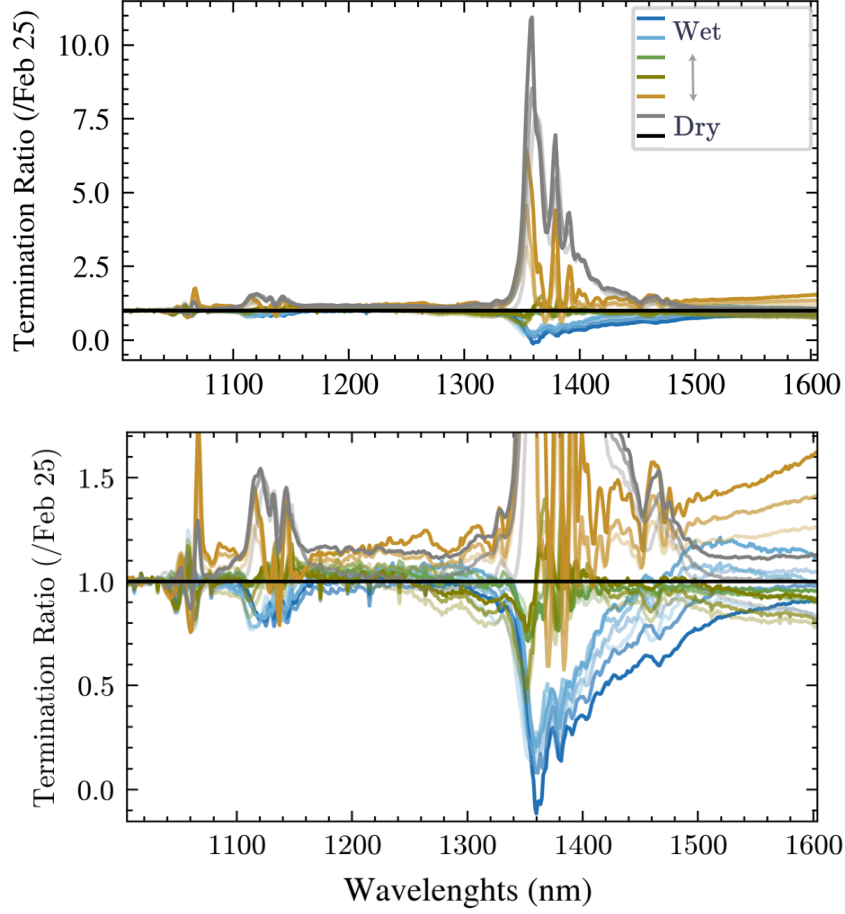


Figure 30: Termination signals divided by that of February 25th to highlight attenuation due to water absorption throughout the beam path.

the opposite trend. This proved that long-range attenuation of water absorption bands could be spectrally resolved.

The strength of the signal of each wavelength used to measure water attenuation was measured along the beam path. This resulted in low SNR signals, particularly for the water vapour absorption line at 1364 nm, shown in Figure 31. From these lines, the log of the ratio between the on-line absorption wavelengths and off-line wavelengths was determined, where the slope was measured to determine the attenuation coefficient. The results for days with high aerosol loading and low aerosol loading, respectively, are shown in Figure 32, where the linear fit is much more stable for higher signal strengths, resulting in more accurate retrieval. After around 70 meters however, a non-linear decrease appears, signifying low SNR.

A wider span of wavelengths was then introduced, where the corresponding termination signals now shown in Figure 33, where the water concentration again resulted in differences in termination strength around the two absorption bands. The DIAL retrieval was performed for these spectral bands, resulting in signif-

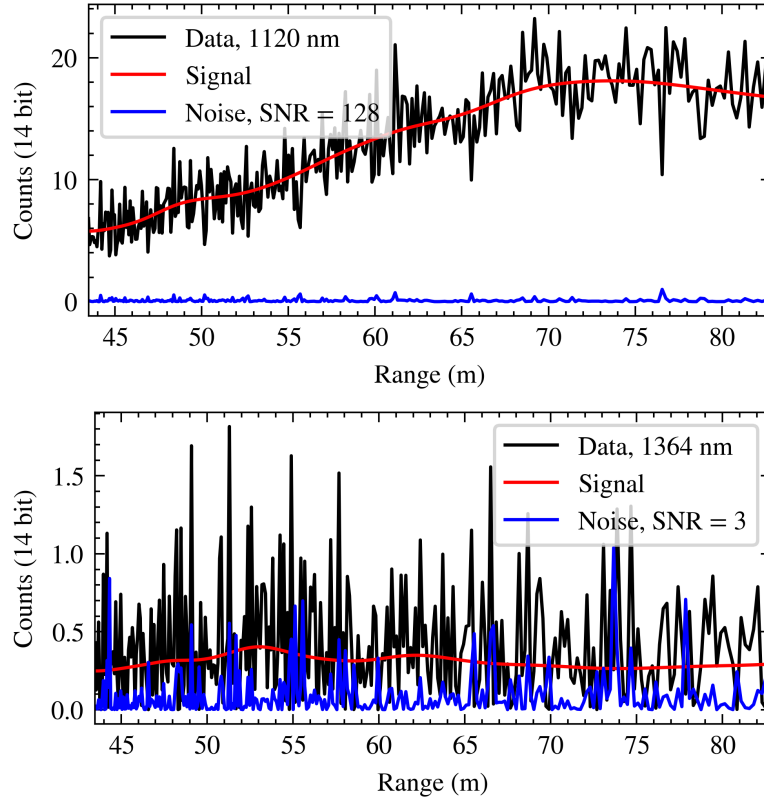


Figure 31: SNR of signals at different wavelengths over the detector range. Signal refers to the smoothed data and noise is the squared difference between data and signal.

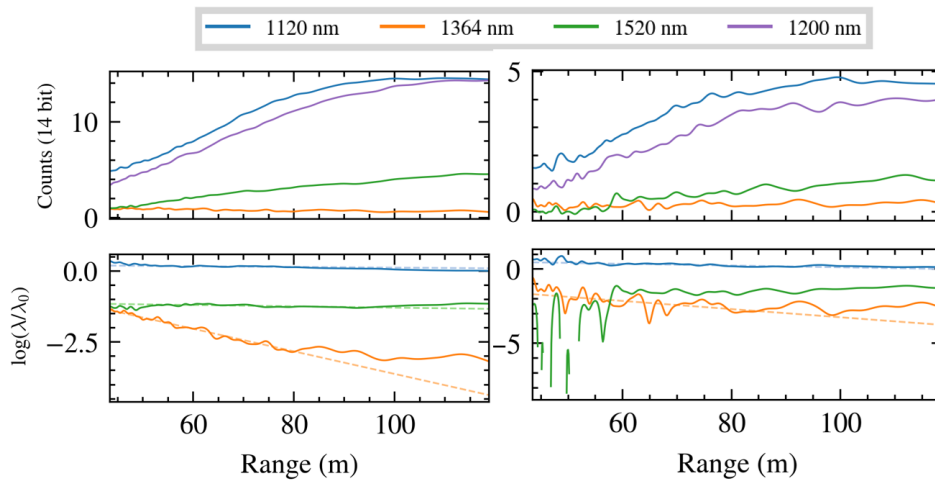


Figure 32: Dial measurement on March 10 (left), March 25 (right), showcasing the reduced SNR resulting from low aerosol loading.

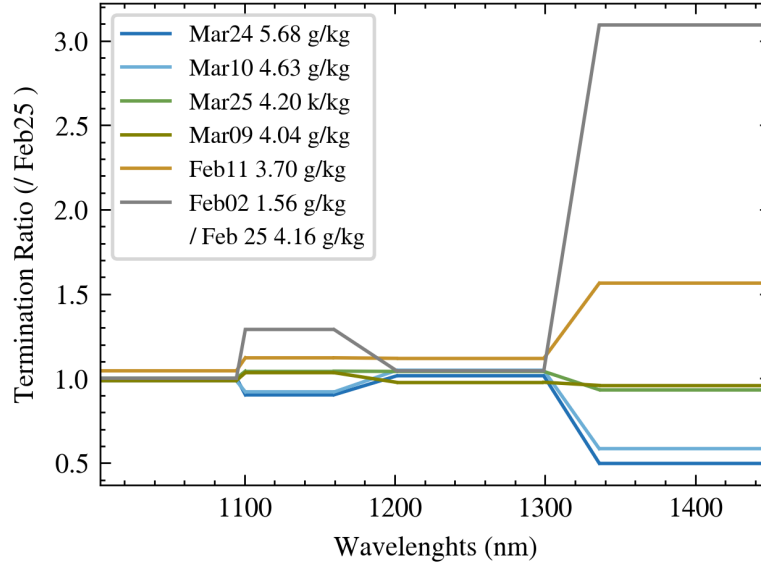


Figure 33: Termination signals of different days divided by that of Mar 25 of averaged bands: 2 off-line and 2 on-line bands.

icant improvements in the linear fits of the absorption coefficients. The results from March 10 and March 25 are shown in Figure 34. Using the mean strength of the absorption bands, the water concentrations were estimated and compared with the Lund ambient water concentrations. The results are shown in Table 2.

Table 2: DIAL retrieval results. Abs 1 signifies the strong absorption band at 1400 nm and Abs 2 the weak one at around 1130 nm. These are compared to the Lund concentrations for the respective day.

Abs 1 (k/kg)	Abs 2 (g/kg)	Lund vals (g/kg)
3.9	5.4	5.7
3.8	4.3	4.6
3.7	8.3	4.2
2.9	3.7	4.0
2.3	9.3	3.7
2.2	6.3	1.6

4.5 Retrieval possibilities

Retrieval algorithms were performed for widths and modes of log-normal distributions of accumulation mode and coarse mode sea salt aerosols. The broadband source in the visible spectrum was most efficient overall to retrieve particle size distributions. Plots of the retrieval processes are shown for the accumulation retrieval in Figure 35.

Table 3 shows the results for the simulated retrievals of mode and width, for the three different wavelength regimes, first for unimodal distributions then for

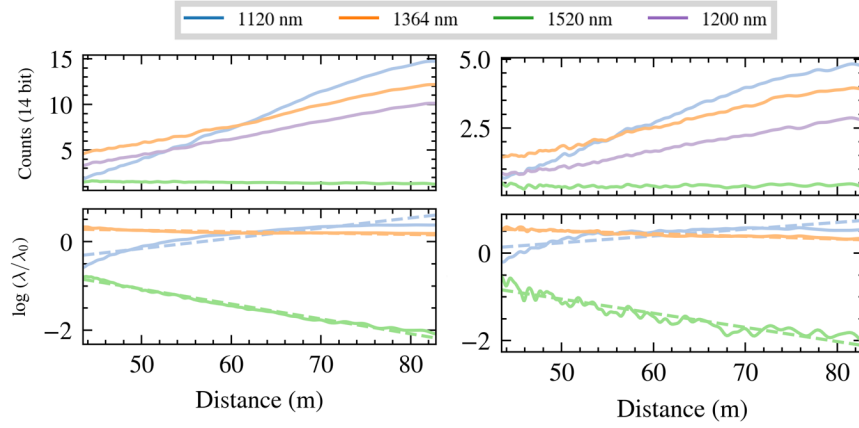


Figure 34: Wider band DIAL measurement on March 10 (left), March 25 (right). Averaging across different bands can be seen to improve the linear fit.

bimodal. Contrary to expectations, all wavelength types performed better in the bimodal retrieval than in the unimodal retrieval. This could be due to higher overall information content, constraining the retrieval.

Overall the 300-1000 nm broadband spectral data performed best in retrieving particle distribution parameters. This showcases the benefits of broadband light sources compared to single wavelength lasers, yielding higher information content. The contrast between the performance of the 300-1000 nm retrieval and the 1000-1600 nm can be caused by the higher wavelength band being large compared to the accumulation mode size, yielding low wavelength depending scattering.

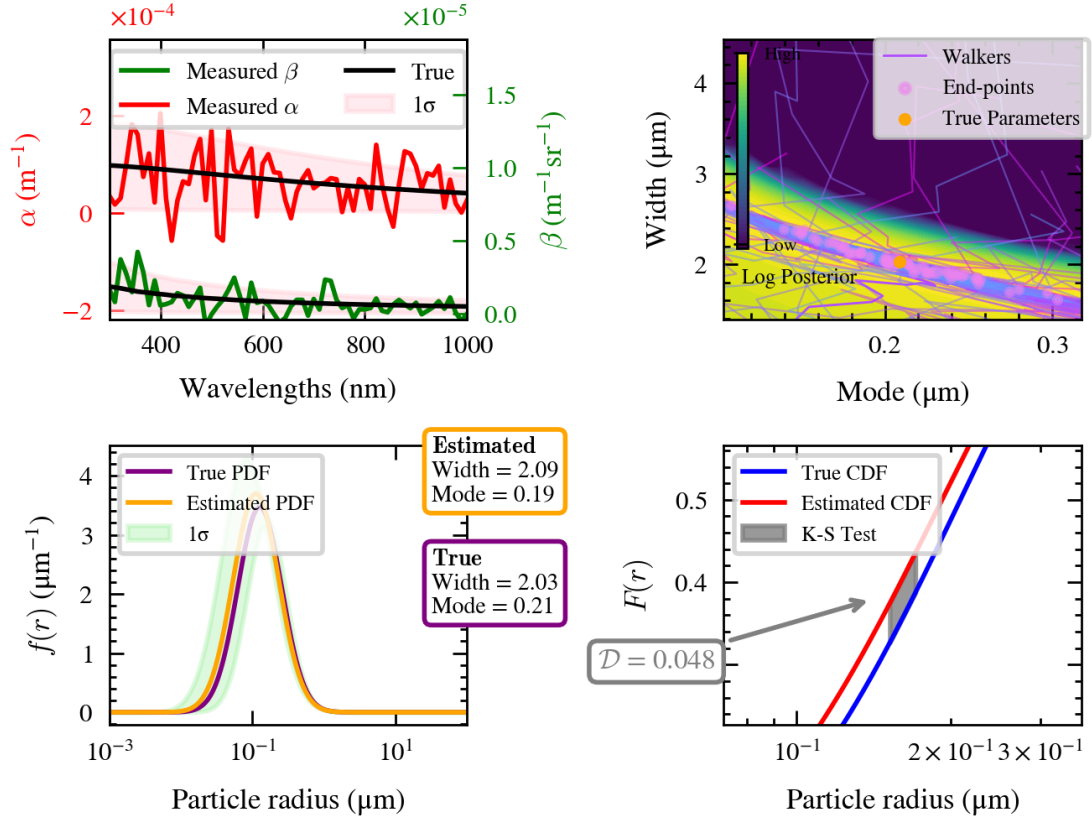


Figure 35: Simulation results of broadband 300 to 1000 nm retrieval of width and mode of particle distribution of water accumulation size mode. (Top left) Sampled extinction and backscatter coefficients with 20 percent noise used to determine the optimal size distribution. (Top right) Monte Carlo walkers exploring parameter space of Width and mode, to optimise log posterior likelihood. (Bottom left) retrieved vs true particle distribution in log scale. (Bottom right) Kolmogorov-Smirnov test of PDFs as a goodness-of-fit measure

Table 3: Performance of broadband wavelength retrievals for different aerosol mode configurations.

Wavelength range	width (σ)	Mode	D-score
Accumulation mode			
True	2.03	0.21	
300-1000 nm	2.09	0.19	0.048
1000-1600 nm	2.59	0.12	0.293
365,532 1064 nm	1.70	0.29	0.230
Coarse mode			
True	2.03	1.75	
300-1000 nm	3.16	0.82	0.360
1000-1600 nm	2.05	1.78	0.122
365,532 1064 nm	2.90	0.91	0.324
Bimodal accumulation mode			
True	2.03	0.21	
300-1000 nm	2.05	0.21	0.008
1000-1600 nm	2.44	0.13	0.257
365,532 1064 nm	1.93	0.23	0.070
Bimodal coarse mode			
True	2.03	1.75	
300-1000 nm	2.2	1.6	0.064
1000-1600 nm	1.88	1.9	0.062
365,532 1064 nm	2.24	1.6	0.079

5 Discussion

5.1 Spectral response of atmospheric targets

The hyperspectral Lidar method has been implemented and has produced spectrally resolved measurements of various atmospheric targets, including precipitation, ambient aerosols, clouds, and aerosol plumes. This showed wavelength-dependent scattering and absorption behaviour. Aerosols produce Mie scattering, dependent on size and refractive index. Water vapour on the other hand showed distinct absorption features. Ice crystals produced specular reflection from their surfaces as well as dispersion effects.

Overall, higher signal strengths yielded richer spectra due to increased signal-to-noise ratio. This was the case with specular reflection from the snowflakes and high aerosol backscattering from fog and plumes. The hyperspectral lidar provided a continuous spectrum of atmospheric information, unlike conventional lidar systems. This allows simultaneous measurements of DIAL absorption across multiple absorption bands, wavelength-dependent aerosol backscatter and extinction coefficients, and probing of cloud microphysics.

5.2 Interpretation

The strongest wavelength dependence was measured from the snowflakes, which produce iridescent features. This was likely due to their hexagonal structure and flat surfaces, acting as dispersive mirrors producing high signals from specular reflection. This explains why radically different signals are produced from each snowflake. As each snowflake is aligned slightly differently and has a different shape and size, different wavelengths are enhanced from the refraction at its surface. The wavelengths produced are most likely specularly reflected back to the receiver at the same wavelength.

Signal strengths were also rather high for fog and aerosol plumes where many scatterers are present, producing a diffuse reflection or backscattering pattern. Aerosol scattering is diffuse, with enhanced forward scattering, leading to a weaker overall signal.

For fog droplets and aerosol plumes, the high signal strength meant that the full emitted spectrum was captured by the receiver and could then be analysed. The lidar ratios determined for the plume releases highlighted this possibility and showed a stable curve for the entire wavelength range. Extinction could also be retrieved as for the snowflakes in different bands using the slope method.

For lower-return measurements, such as ambient aerosols, fewer counts indicated that the signal was not distributed across the entire spectrum. Using the mean signal, it was still possible to measure cloud-like dust clouds from the road, as well as wind direction and speed. The low returns are especially true for the large absorption line at 1400 nm, where high extinction was present, also present in the

DIAL measurements, leading to low SNR.

For optically dense media, such as fog and plumes, a decrease in the termination signal was generally observed, as expected with increasing optical depth, consistent with Beer-Lambert attenuation. In the fog data, there is a deviation from this where termination and fog signals were positively correlated. This is likely due to multiple scattering, in which photons are scattered multiple times within the cloud. If the angle of the multiple scattered photons is the same as that of the termination, this signal will appear as if it were from the termination. This effect diminishes as optical depths become higher which could be due to high extinction at low distances, further from the termination, not yielding the same scattering effect.

For plumes, lidar ratio was possible to determine, at around 30 - 50 sr. This is typical for atmospheric aerosols. Backscatter and extinction do not seem to have a linear correlation, where backscatter saturates at high optical depths. This is similar to the fog data; however, the typical lidar ratios retrieved highlighted that multiple scattering was not overwhelmingly present. However, this would likely have broadened the backscattered light, along with the wide particle distributions in the fog.

5.3 Limitations

5.3.1 Optical Depth and Multiple Scattering

The high optical depth along the path for fog and plumes implied multiple scattering. This breaks down Beer-Lambert assumptions for signal attenuation and changes the effective backscatter of the aerosols. This limited the retrieval of lidar ratio, broadened the spectrum, and reduced the range resolution of the system. Broadening the spectrum reduced the effective wavelength dependence of the scattering, reducing retrieval possibility of microphysical properties [24]. The reduced range resolution comes from the larger spatial distribution of the backscattered light, such that signal strength increases at nearby pixels as well. This increases the termination signal for example, which would reduce the measured optical depth. At high optical depth, saturation also occurred, reducing the apparent signal.

5.3.2 Particle distributions and spectral dependence

Particle distributions at larger radii become increasingly spread out in space, which smoothens Mie oscillations and wavelength dependence of the scattering. This resulted in the spectra not changing much in values over the spectral range. This reflected the difficulty in producing constrained particle distributions. In the measurements, fog and mechanically generated aerosols, as well as ambient PM10 particles, are also larger than the wavelengths, which implies geometric scattering.

5.3.3 Signal to noise

The limitation of spreading the signal over multiple wavelengths is that the signal strength reduces correspondingly. This lowers the signal count and more averaging has to be done to obtain good photon statistics over the entire wavelength range. The large particle distributions also yielded higher forward scattering than the 1-2 micron Mie scattering, reducing the signal. The SNR was further reduced by detector heating and dark current that could not be fully removed. The spectrum not being uniform meant that a lower signal-to-noise ratio resulted in calibration becoming more difficult. Overall, low pollution levels, especially at PM₁, PM_{2.5} meant that the SNR of retrieval was reduced.

5.3.4 Retrieval and calibration limitations

Due to the non-uniform spectrum, low SNR and multiple scattering combined, the calibration for backscatter and extinction coefficients were difficult to retrieve and was spectrally flat. From the simulations, this is probably due to the large width of the particle size distribution range not yielding many features in the SWIR spectral range. Also, since the particle distributions were not constrained, the retrieval of size distributions was not possible. Lastly, since neoprene was needed for backscatter retrieval, this was not possible for atmospheric measurements.

The simplifying assumptions of Mie theory are always a limitation when treating aerosols, as uncertainties are introduced by non-sphericity and non-lognormality. This was clear in snowflakes, whose reflections were most likely not due to Mie scattering.

5.4 Evaluation

The hyperspectral Lidar signal was shown to have the possibility of providing high-resolution measurements in time, range, and wavelengths. Low-SNR signals, such as those from ambient aerosols, yielded lower spectral resolution, while high-SNR signals, such as those from plumes, fog, and snow, yielded higher spectral resolution.

The method proved successful in the analysis of signals and determining Lidar ratios, optical depths, and molecular absorption for multiple wavelengths.

The limitation of not obtaining a high backscatter signal while maintaining Mie assumptions were underestimated. This was enhanced by the difficulty in producing a narrow particle size distribution, which meant that the tasks of determining aerosol specifics were more difficult than originally expected.

5.5 Improvements

To improve the spectral retrieval of atmospheric constituents, a first step is to increase the SNR. This could be done by cooling the detector to reduce temperature impact on dark current. This would reduce the non-linear increase in the signal across the detector and enhance it. This would also enable one to increase the exposure time, thereby increasing the number of counts measured, without having to account for drifting dark current. This would allow for better calibration of the backscatter signal.

To enhance spectral signatures in the Mie regime, aerosols generated within a narrow range around the emitted wavelengths would need to be used. This could be done by using monodisperse lab-generated aerosols. A spectrum in the UV or visible would also be favoured as particle distributions at those sizes are naturally narrower.

The constraints on the experiment could then be overcome by using an optical particle counter (OPC) or other in-situ measurements alongside the Lidar signal. To reduce the influence of ambient atmospheric conditions, a controlled lab environment could also be used. This would also allow the investigation of controlled bi-modal distributions to measure the potential of inversion techniques. A monitoring camera, as well as a weather station, could also be useful for determining ambient conditions and monitoring oncoming traffic and dust. A more constrained environment could be achieved by measuring outside an urban area, where there is less variability along the beam.

To enhance the resolution of the system, to enable measurements at larger distances, the baseline between the receiver and emitter could be extended. Moreover, released aerosols could be scattered throughout the entire beam to enable range-dependent backscatter and extinction coefficients, using Klett inversion. This would also reduce multiple scattering.

To better measure the capability of the system and to collect more data, longer time-scale measurements could be done to measure the sensitivity to determine variations in atmospheric conditions and air quality over time.

6 Outlook

The effect of aerosols on radiative forcing remains an uncertainty in current climate models. This requires new methods to be developed to better understand their concentrations and radiative effects in the atmosphere. By introducing more wavelengths, aerosol scattering contributions can be better constrained.

In this thesis, we have introduced the Scheimpflug hyperspectral lidar method, to retrieve spectral signatures of different atmospheric constituents. The method was tested by probing different atmospheric optical regimes.

Hyperspectral measurements were successfully made for multiple atmospheric targets. It was found that high signal strengths generally corresponded to high spectral information, especially for dispersive ice crystals. Optical quantities such as backscatter and extinction coefficients could be determined while resolving oscillatory Mie scattering proved to be difficult to obtain in the current setup. It was found however that even for relatively low signals, molecular absorption could be quantified.

Characterising multiple atmospheric constituents helps provide a better overall understanding of the atmospheric composition. Information from molecular, aerosol, and larger atmospheric constituents can be combined to better constrain radiative transfer effects in the atmosphere. This includes direct scattering and absorption from the particles but also their combined effect on cloud formation and albedo.

The main benefit of this method is that all of this can be done simultaneously in a single measurement. This ensures cross-calibration and atmospheric consistency between measurements.

The compactness of the Scheimpflug setup, low-maintenance and portability allow for long-term measurements and reduced costs. This is ideal for satellite missions, where one could develop this method further to enable a unified atmospheric characterisation.

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