



Laboratory setup to study the polarization properties of size-resolved aerosol particles in exact back-scattering direction

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Abstract. In this study, a new laboratory-based experimental setup, the Optical Lab for Lidar Applications (OLALA), was developed and characterized to measure the polarization properties of size-resolved aerosol particles in the exact backscattering direction. The linear depolarization ratio was measured at 532 nm wavelength at a scattering angle of $180 \pm 0.2^\circ$ to be comparable to lidar observations. To realize exact backscattering measurements, a 50:50 beam splitter was used to establish a
5 coaxial transmit-receive optical configuration. Aerosols were generated using an atomizer and size-selected with a differential mobility analyzer, which allowed to measure particle size distributions in the fine mode (mostly below 1 μm). Polystyrene latex (PSL) beads, ammonium sulfate, sodium chloride, Arizona Test Dust (ATD), and Saxonian soil dust were used as test
10 aerosols to evaluate the performance of the system. PSL particles of diameter 1 μm exhibited the lowest depolarization ratio of 0.017 ± 0.002 , which agrees with theoretical values for spherical particles. Arizona Test Dust, which is characterized by an
irregular particle shape, produced the highest depolarization ratio of 0.168 ± 0.048 for particles having a median diameter of 766 nm. For the non-spherical particles like sodium chloride, ATD, and Saxonian soil dust, an increase of the depolarization ratio with increasing size was observed.

1 Introduction

Aerosol particles interact with solar and terrestrial radiation and influence cloud and precipitation formation. They are highly
15 variable in time and space. Thus, continuous monitoring is desired. Lidar (light detection and ranging) instruments are well-suited to provide continuous, vertically resolved observations of aerosol particles. The lidar-observed optical properties characterize well the particle type, such as mineral dust, urban pollution, smoke, or marine aerosol (Burton et al., 2012; Groß et al., 2013; Floutsi et al., 2023; Wandinger et al., 2023). In particular, the depolarization ratio is of special importance because it indicates the presence of non-spherical particles such as mineral dust (e.g., Freudenthaler et al., 2009; Haarig et al., 2017a), dry
20 sea salt (Haarig et al., 2017b; Ferrare et al., 2023), stratospheric smoke (Haarig et al., 2018; Hu et al., 2019) and ice crystals (Sassen, 1991; Kustova et al., 2022). The irregular shape of these particles challenges a proper description in scattering theory, as the often-used Mie theory does not apply to non-spherical particles. Here, mineral dust is of particular interest because of its high abundance in the atmosphere (Chooari et al., 2014). The description of its highly irregular shape characteristics has received a lot of attention in recent years (e.g., Chang et al., 2025; La Luna et al., 2025). Several particle shape models



25 were proposed, such as spheroids (Dubovik et al., 2006), super-spheroids (Bi et al., 2018), deformed spheroids (Kahnert et al., 2020), irregularly shaped particles (Gasteiger et al., 2011) or irregular hexahedra (Saito et al., 2021). Most of these models are evaluated against laboratory measurements, e.g., from the Amsterdam – Granada light scattering database (Volten et al., 2001; Muñoz et al., 2012, 2025) which covers the range of scattering angles from 3 to 177°. This angular range is generally sufficient for many passive remote sensing applications. For example, the Aerosol Robotic Network (AERONET) sky-radiance retrievals
30 typically use only a subset of the full scattering angle range, depending on the observation geometry (Dubovik and King, 2000). However, for the application to lidar measurements, observations at a scattering angle of 180° are required because lidar retrievals depend on the backscattering phase function, and the extrapolation to the exact backscattering direction is not always trivial.

First laboratory measurements of the depolarization ratio were performed already in the 20th century by Cooper et al. (1974)
35 and Sassen et al. (1989), who worked with sea salt and sulfuric acid, respectively. Pioneering laboratory work in the field of dust research was performed by Cao et al. (2010) and Sakai et al. (2010), who worked with lidar-like laboratory setups, i.e., they covered scattering angles of almost 180°. Especially, the separate measurement of the depolarization ratio of fine and coarse mode dust was a significant achievement by Sakai et al. (2010) that greatly supported the development of dust separation algorithms from polarization lidar measurements (Mamouri and Ansmann, 2014; Shang et al., 2026). Järvinen et al.
40 (2016) observed the depolarization ratio of different mineral dust particles with broad mono-modal particle size distributions at a scattering angle of 178° and could clearly show an increase of the depolarization ratio (at 552 nm) up to a mean volume median diameter of around 1.25 µm. Then, the depolarization ratio plateaued for larger sizes at a value of around 0.3. The setup of Gautam and Sorensen (2021) covered a wide range of sideward scattering angles (0.32 – 177.6°) with strongly enhanced observation capabilities in the forward scattering direction. Martikainen et al. (2024) did measurements of size-selected Martian
45 dust analogs over a scattering angle range from 3° to 177° at 488 and 640 nm. The research group in Lyon reached the exact backscatter (180±0.2°) direction (David et al., 2013). Further improvements to their setup included a dual-wavelength setup at 355 and 532 nm (Miffre et al., 2016), observations from near to exact backscatter (Miffre et al., 2019), and the observations of pure minerals such as quartz and hematite (Miffre et al., 2023).

Building on the experiences of previous laboratory setups and our own experience in lidar design (e.g., Althausen et al., 2000;
50 Engelmann et al., 2016), we present a novel experimental setup called the Optical Lab for Lidar Applications (OLALA) in Section 2. In this scattering laboratory, we observe the depolarization ratio in the exact backscattering (180 ± 0.2°) of size-selected aerosol samples. In this first study, we present the design of the particle generation and size selection unit (Sections 2.2 and 2.3), and the optical setup at a wavelength of 532 nm for achieving the 180° scattering angle (Sect. 2.4). The extension of the optical setup to include the wavelengths of 1064 and 355 nm is in preparation. To prove the concept and functionality of our
55 new laboratory, we tested it with known spherical particles such as polystyrene latex (PSL) beads, with size-selected sodium chloride and ammonium sulfate particles generated from solutions, and then used more complex particles, which are commercially available, namely Arizona Test Dust (Sect. 3). Finally, we introduce some natural soil dust from Saxony (Germany) to demonstrate the ability of the laboratory to use real-world dust samples – a path which we will follow in the future with dust



samples from various deserts on Earth. In Sec. 3.2, we present the comparison of our laboratory measurements against the modeled values. In Section 4, we conclude our findings and provide an outlook on future activities of OLALA.

2 Instrumentation – the new scattering laboratory OLALA

A new scattering laboratory was established at the Leibniz Institute for Tropospheric Research (TROPOS). The facility, named OLALA was developed to create a comprehensive dataset of scattering properties of size resolved aerosol particles in the exact backscattering direction, relevant for lidar applications. OLALA combines aerosol generation, particle size selection and polarization resolved optical measurements to determine the backscattering properties of well characterized aerosol particles. In the following section, we present a detailed description of the used aerosol samples (Sect. 2.1), the experimental setup, including the particle generation (Sect. 2.2), size selection (Sect. 2.3), and the current optical setup operating at 532 nm (Sect. 2.4). Furthermore, we describe the experimental procedure (Sect. 2.5) and estimate the related uncertainties (Sect. 2.6).

2.1 Aerosol samples

We used a variety of aerosol particle types to cover a diverse range of morphologies, including spherical, cubic, and irregularly shaped particles, to evaluate the performance and sensitivity of our experimental setup. For this study, the aerosol particles were prepared from wet dispersions of their samples. A brief description of all the aerosol samples used in this study is given below.

– Polystyrene Latex (PSL) particles

PSL particles are available in a range of well-characterized diameters and exhibit a uniform and narrow size distribution. Due to their precisely defined physical properties and spherical morphology, they were used as reference particles to evaluate the performance of our experimental setup for perfectly spherical particles. To produce aerosol of PSL, some droplets of PSL particles (Duke scientific) were added to ultrapure water in a medical droplet disperser (MEDAP MA-QUET hand-held nebulizer) that replaced the aerosol generator mentioned in section 2.2. The refractive index of the PSL particles was $1.615 + 0.001i$ at $495 \text{ nm} \pm 5 \text{ nm}$ as provided by the manufacturer.

– Ammonium Sulfate ($(\text{NH}_4)_2\text{SO}_4$)

Ammonium sulfate particles have a spherical morphology similar to PSL particles. However, in contrast to PSL particles, ammonium sulfate is not available in well-defined, monodisperse sizes. It is, however, highly soluble in water. Therefore, ammonium sulfate aerosol particles were generated using the setup described in section 2.2 from an aqueous solution of ammonium sulfate (Fluka, > 99.5%; CAS RN 7783-20-2) prepared in ultrapure water, with an ammonium sulfate concentration of 5 g/L. Particle size selection was carried out as part of the experimental procedure using a Differential Mobility Analyzer (DMA) as described in section 2.2. The refractive index of ammonium sulfate is 1.53 in the visible spectral region (Toon et al., 1976).



– Sodium Chloride (NaCl)

Sodium Chloride particles are generally non-spherical but have a less complex structure compared to mineral dust particles. Under low-humidity atmospheric conditions, NaCl crystals effloresce from NaCl-solution droplets. These crystalline particles are predominantly cubic in shape with rounded edges. Even for atmospheric particles generated from sea spray, a cubic shape was observed (Haarig et al., 2017b). Due to their well-defined shape, they were used to examine the impact of non-spherical but geometrically regular particles on the measured optical parameters and could be compared to optical calculations (Sect. 3.2). The NaCl particles were generated in a similar way as ammonium sulfate (using NaCl from Sigma Aldrich, > 99.5%; CAS RN 7647-14-5). The refractive index of sodium chloride is $1.5663 + 0.0i$ at a wavelength of 405 nm (Eldridge and Palik, 1985).

– Arizona Test Dust (ATD)

ATD (ISO 12103-1, A1 Ultrafine Test Dust; Powder Technology Inc.) was used as a standardized mineral dust with irregular particle morphology. It consists mainly of silicate minerals such as quartz and feldspar, and was used as an analogue for natural mineral dust aerosols. The ATD aerosols were generated from suspensions in a similar way as ammonium sulfate and NaCl. Only an additional magnetic stirrer was used to constantly stir the suspension and prevent particle settling during the experiments. Miffre et al. (2016) measured a complex refractive index for ATD of $1.55 + 0.005i$ at 532 nm.

– Saxonian Soil Dust

In addition to commercially available samples, we also did measurements with natural soil dust. The soil dust sample was collected from an agricultural area near the town of Grimma in Saxony, Germany (51.243°N, 12.635°E) from a depth of about 30 cm. The sample was dried for three days at 121°C and sieved to remove particles larger than 100 µm. To generate aerosols of natural soil dust, the soil sample was suspended in deionized water and was left to settle overnight. The supernatant liquid containing particles < 5 µm was then used further for particle generation using the same procedure as for ATD.

2.2 Particle generation and aerosol tube

The steps followed for particle generation, size selection, and measurement are shown in a sketch in Fig. 1 (a). An aerosol generator (design similar to TSI 3076) was deployed to produce aerosols from wet dispersion by spraying the samples from solutions/suspensions in water. After spraying, the aerosol was dried in a diffusion dryer to eliminate excess water, leaving it with a relative humidity (RH) of less than 10%. The freshly produced polydisperse aerosol was first passed through a neutralizer to establish a charge distribution on the particles. Then, it was introduced into a Differential Mobility Analyzer (DMA, type "Vienna medium", Knutson and Whitby, 1975) for size selection. The DMA separates the particles by passing them through an electric field, where only particles of specific electrical mobility (roughly equivalent to size) can exit. This typically produces a quasi-monodisperse aerosol population with a narrow mobility diameter spread of approximately 5%. However, very high



concentrations were needed in the samples sprayed by the atomizer (5 g material per liter of solution / suspension) to produce a sufficient amount of particles for the optical measurements. These high particle concentrations reduced the effectiveness of the particle size classification, resulting in broader size distributions. Therefore, a Portable Optical Particle Spectrometer (POPS, Handix Scientific Inc.) was used to characterize all resulting particle size distributions (described in Section 2.3) for the optical measurements done in this study.

After size selection, the aerosol particles enter the aerosol tube, which is 1 m long, has an inner diameter of 1.7 cm, and is vertically mounted (see Fig. 1 (c)). The top of the aerosol tube was closed with a beam dump, and all inner surfaces of the tube were coated with matte black paint to minimize stray light caused by internal reflections. During the experiment, aerosol particles were transported through the aerosol tube by a laminar flow of 1 L min^{-1} . The flow of aerosol particles into the tube was regulated by a remote controlled automatic pneumatic valve, which changed the flow of particles at regular intervals of 10 s, such that the aerosol particles either entered the tube directly or via a filter, filtering out all particles so that only particle-free air entered the tube. The meaning and operation of this switching procedure is described in detail in the experimental procedure (Sect. 2.5).

The number concentration of size-selected aerosol particles at both the inlet and outlet of the aerosol tube could be measured using a Condensation Particle Counter (CPC 3010, TSI). The CPC directly behind the DMA was used before the beginning of the experiment to monitor the generated particle concentrations. This CPC was switched off once the aerosol was fed into the tube, and the CPC at the outlet of the tube was then used to enable optical inspection of the aerosol passing through the tube throughout the experiment.

2.3 Size distribution

The size distribution of each DMA-selected sample was measured using the POPS instrument. As mentioned above, this characterization was done as broad size distributions resulted from the particle generation. The Differential Mobility Analyzer (DMA) used in our experiment selects particles according to their electrical mobility, which depends on both particle size and charge. Because electrical mobility increases with increasing charge, larger particles carrying multiple charges can have the same mobility as smaller singly charged particles (Wiedensohler, 1988). Consequently, in addition to singly charged particles, the selected aerosol also contains larger particles. For a selected size of 250 nm (mobility diameter), doubly, triply, and quadruply charged particles have sizes of 414 nm, 571 nm and 725 nm (mobility diameter), respectively. Additionally, supermicrometer particles can be highly charged, a state that is difficult to determine. High concentrations in the solutions / suspensions that needed to be used in this study, together with multiply charged particles, lead to the observed broad size distributions measured with POPS.

The POPS instrument used in this study operates at 405 nm wavelength and was calibrated with PSL particles of diameter $495 \text{ nm} \pm 5 \text{ nm}$ and refractive index of $1.615 + 0.001i$. POPS measurements were performed for four DMA-selected size fractions named A, B, C, and D, corresponding to mobility diameters of 250 nm, 450 nm, 650 nm and 800 nm, respectively, for each aerosol type. Figure 2 shows the size distribution observed with POPS for four DMA-selected size fractions of ammonium sulfate, sodium chloride, Arizona Test Dust, and Saxonian soil dust. POPS determines particle sizes based on light scattering

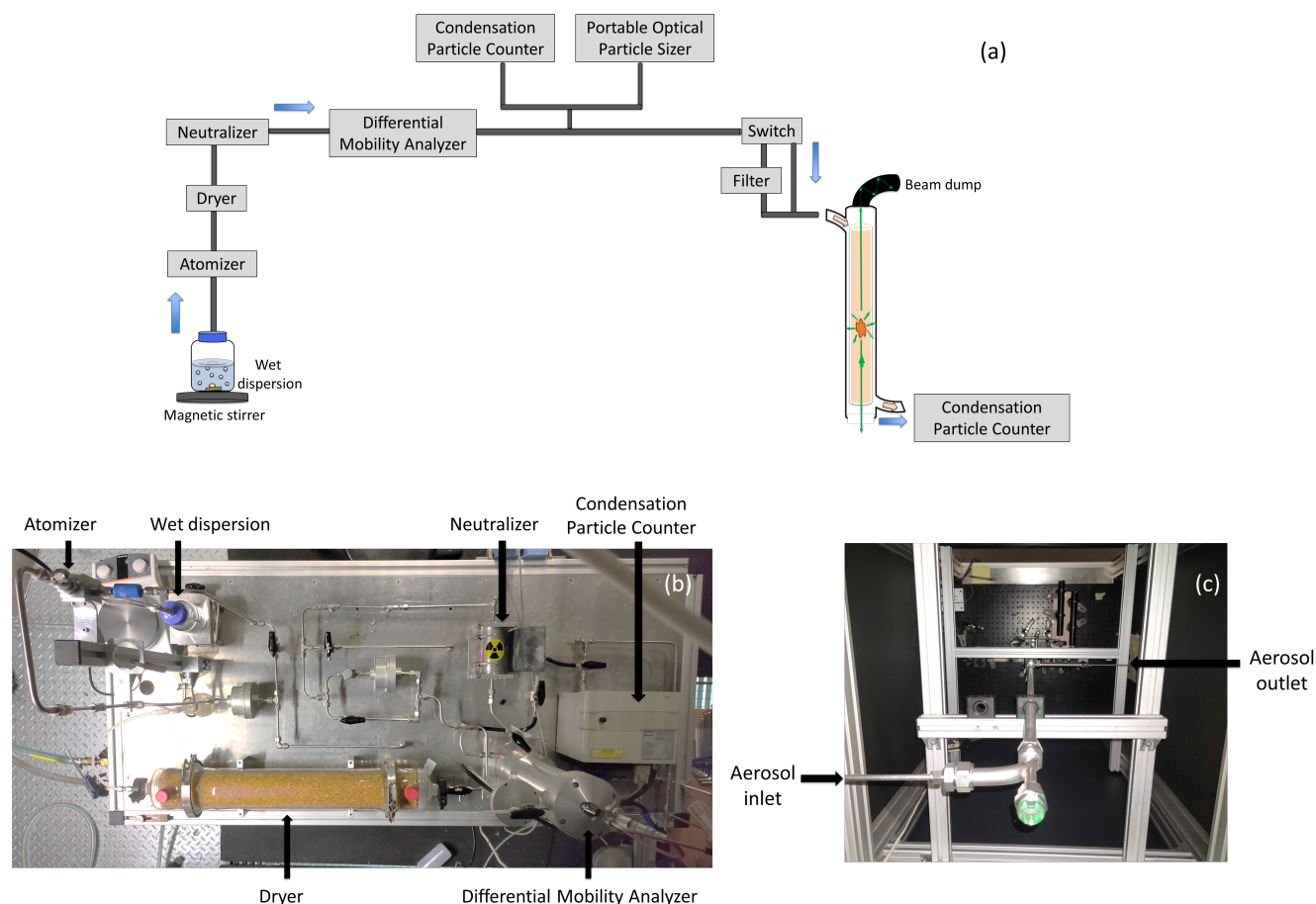


Figure 1. The setup of OLALA: (a) a simple sketch to show the sequence from particle generation to measurement. (b) shows the top-view of the particle-generation and size-selection unit in the laboratory. (c) shows the top view of the vertically mounted 1 m long aerosol tube (with the beam dump on top being removed). The optical table is visible at the bottom.

155 by single particles, and it reports a PSL-equivalent optical diameter. For each DMA-selected aerosol fraction, parameters of the number and surface-area-based size distributions measured by the POPS are presented in Table 1. For each distribution, the cumulative distribution function was calculated, and the 25th percentile diameter ($D_{25\%}$), median diameter (D_{med}), and the 75th percentile diameter ($D_{75\%}$) were determined. The interquartile range ($IQR = D_{75\%} - D_{25\%}$) was used as a measure of distribution width. Quantile-based statistics were selected as the primary descriptors of the particle size because several

160 aerosol populations exhibited broad and asymmetric distributions that could not be consistently represented by a single fitted distribution. Therefore, the median was used as the representative particle diameter. All data on size distributions are provided (see the data availability statement).

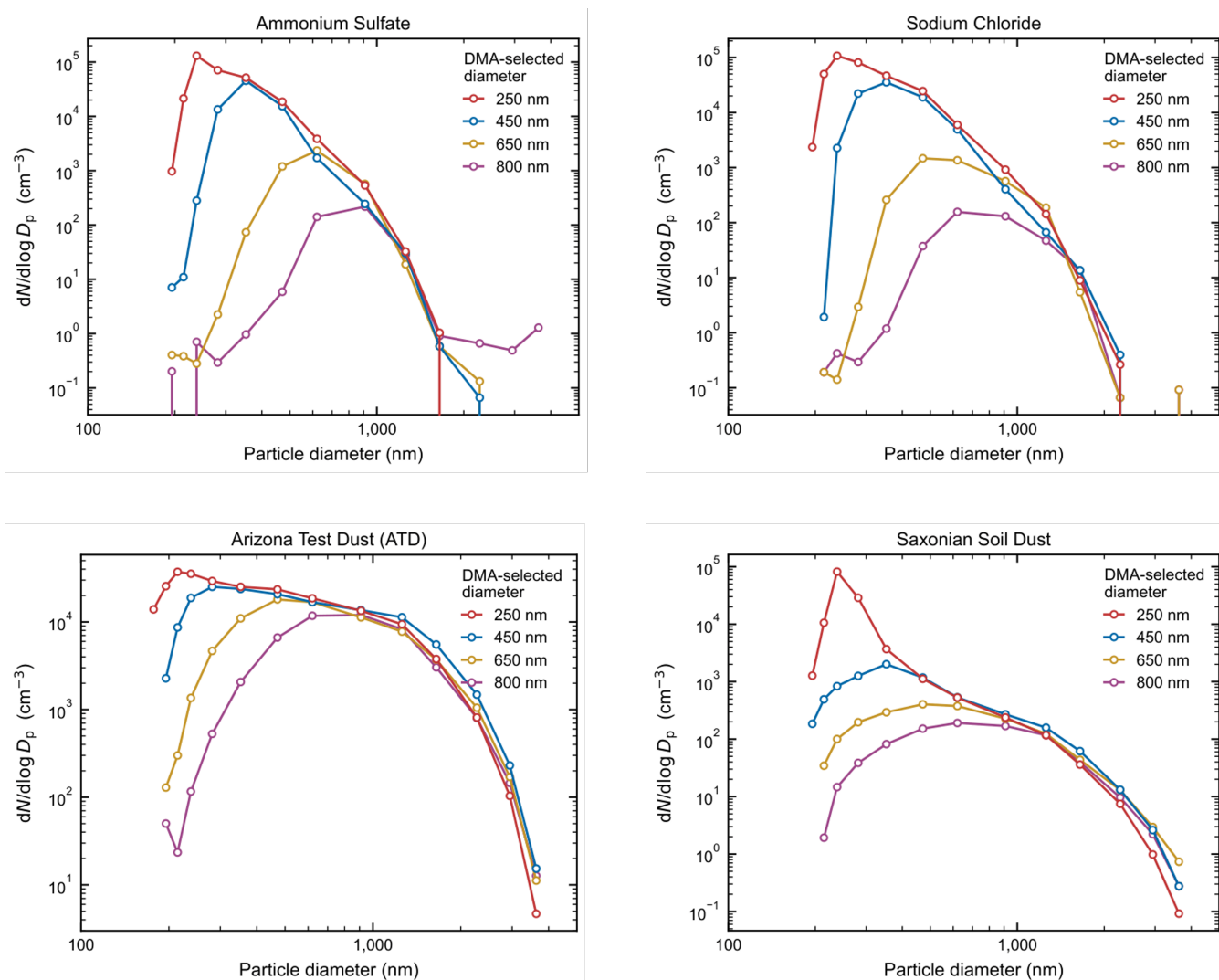


Figure 2. Size distributions as measured with POPS (i.e., optical diameters) for each DMA-selected aerosol size indicated in different colors.



Table 1. Parameters of size distributions as measured with POPS for each DMA-selected aerosol size, including the median diameter D_{med} and the inter-quartile ranges.

Aerosol type	DMA-selected diameter (label)	Number size distribution			Surface size distribution		
		D_{med} (nm)	$D_{25\%}$ (nm)	$D_{75\%}$ (nm)	D_{med} (nm)	$D_{25\%}$ (nm)	$D_{75\%}$ (nm)
Ammonium sulfate	250 nm (A)	281	241	351	341	268	445
	450 nm (B)	360	326	399	382	340	478
	650 nm (C)	606	537	668	642	576	783
	800 nm (D)	798	646	988	913	737	1089
Sodium chloride	250 nm (A)	284	241	359	359	277	495
	450 nm (B)	363	316	449	423	347	537
	650 nm (C)	583	478	679	683	574	1022
	800 nm (D)	699	605	998	975	701	1247
Arizona test dust	250 nm (A)	375	247	630	1094	625	1378
	450 nm (B)	485	318	800	1221	747	1606
	650 nm (C)	602	442	926	1187	713	1570
	800 nm (D)	766	586	1132	1210	862	1525
Saxonian soil dust	250 nm (A)	245	233	276	263	238	330
	450 nm (B)	371	305	518	682	438	1247
	650 nm (C)	555	387	738	1027	636	1362
	800 nm (D)	660	503	1041	1178	789	1444

2.4 Optical setup

- 165 The optical setup resembles the main optical parts of a polarization lidar and consists of an emitter, receiver, and data acquisition. A schematic sketch is shown in Fig. 3. The 180° scattering angle is achieved with the help of a 50:50 beam splitter setup similar to the setup implemented in the Manchester Ice Cloud Chamber (Smith et al., 2016). In the emitter section, we use a diode-pumped fiber-pigtailed continuous wave laser as the light source, operating at the wavelength of 532 nm (Cobolt, Huebner Photonics). The laser fiber output is connected with a FC/APC (Ferrule connector/Angled physical contact) fiber collimator of a numerical aperture of 0.51 and a focal length of 7.86 mm (F240APC-532, Thorlabs). A linear polarizer (LPVISA050-MP2, Thorlabs), placed at the collimator's output, is used to set the polarization of the emitted laser beam. The linearly polarized laser beam is then directed inside the aerosol tube using a 50:50 beam splitter. The 50:50 beam splitter reflects half of the emitted laser beam inside the aerosol tube, and the other half of the incident light is transmitted to a beam dump placed on the optical table.
- 175 The light scattered back by the aerosol particles within the aerosol tube passes through the 50:50 beam splitter once more



before being directed to the receiver section.

In the receiver section, 50% of backscattered light is collected by a dielectric mirror M1 (BB1-E02, Thorlabs) placed just below the 50:50 beam splitter. This dielectric mirror reflects the back-scattered light at an angle of 45° towards the field stop. The diameter of the illuminated volume in the aerosol tube is 2 mm, and the distance between the center of the illuminated volume and the field stop is 830 mm. The diameter of the field stop is restricted to 3 mm, to ensure exact backscattering measurements. Within the effective scattering volume, the angular deviation from 180° remained below $\pm 0.2^\circ$ and reached approximately $\pm 0.1^\circ$ at the center of the aerosol tube. A polarizing beam splitter cube (PBS, PBS251/M, Thorlabs) is placed after the field stop to separate the co-polarized (p) and cross-polarized (s) components of the backscattered light with respect to the polarization plane of the emitted laser beam. The two orthogonal components of the backscattered light are then detected in separate detection units placed at equal distances from the field stop. A cleaning polarizer (LPVISA050-MP2, Thorlabs) is placed in front of each detection unit to suppress unwanted polarization components (cleaned polarizing beam splitter cube according to Freudenthaler, 2016). Each detection unit consists of an interference filter (FLH532-4, Thorlabs) of central wavelength 532 nm and full width at half maximum of 4 nm, a plano convex lens L1 (LA1509-A, Thorlabs) of focal length 100 mm followed by a second pinhole of diameter 0.5 mm. The second pinhole is used as a spatial filter to optimize the ratio between the backscattered light from the volume under investigation inside the aerosol tube and the backscattered (stray) light from the components at shorter distances. After the second pinhole, a plano concave lens L2 (LC1715-A, Thorlabs) of focal length 50 mm is placed to diverge the backscattered light on the photocathode of the photomultiplier tube (H5783, Hamamatsu). All optical components in each of the detection units are put within a housing to minimize stray light. A dual channel lock-in amplifier (SE1022D, Saluki Technology) with a 24-bit analog to digital converter was utilized to detect the backscattered signal from the noisy background and acquire the PMT output of both channels simultaneously. The lock-in amplifier works on the principle of phase-sensitive detection and extracts only the component of the input signal that oscillates at the same frequency as the reference signal. To suppress background noise, two beam dumps were placed in the setup. These beam dumps are small, curved rubber pipes with a black reflective inner surface and one end sealed with black velvet (Acktar Ltd.). The curved specular inner surface of the beam dumps attenuates the backscattered light through successive forward reflections of the incident laser beam. The first beam dump was positioned on the optical table to collect the transmitted light from the 50:50 beam splitter in the emitter section. The second beam dump was used to seal the top of the aerosol tube and to ensure air tightness. The $\Delta 90^\circ$ -method (Freudenthaler, 2016) was implemented to take into consideration the different gains of the two detection channels with the help of a half-wave plate (WPH10ME-532, Thorlabs) placed in front of the PBS. The complete description of the calibration routine is given in the next section.

2.5 Experimental procedure

The techniques and steps followed during the experiments are described below.

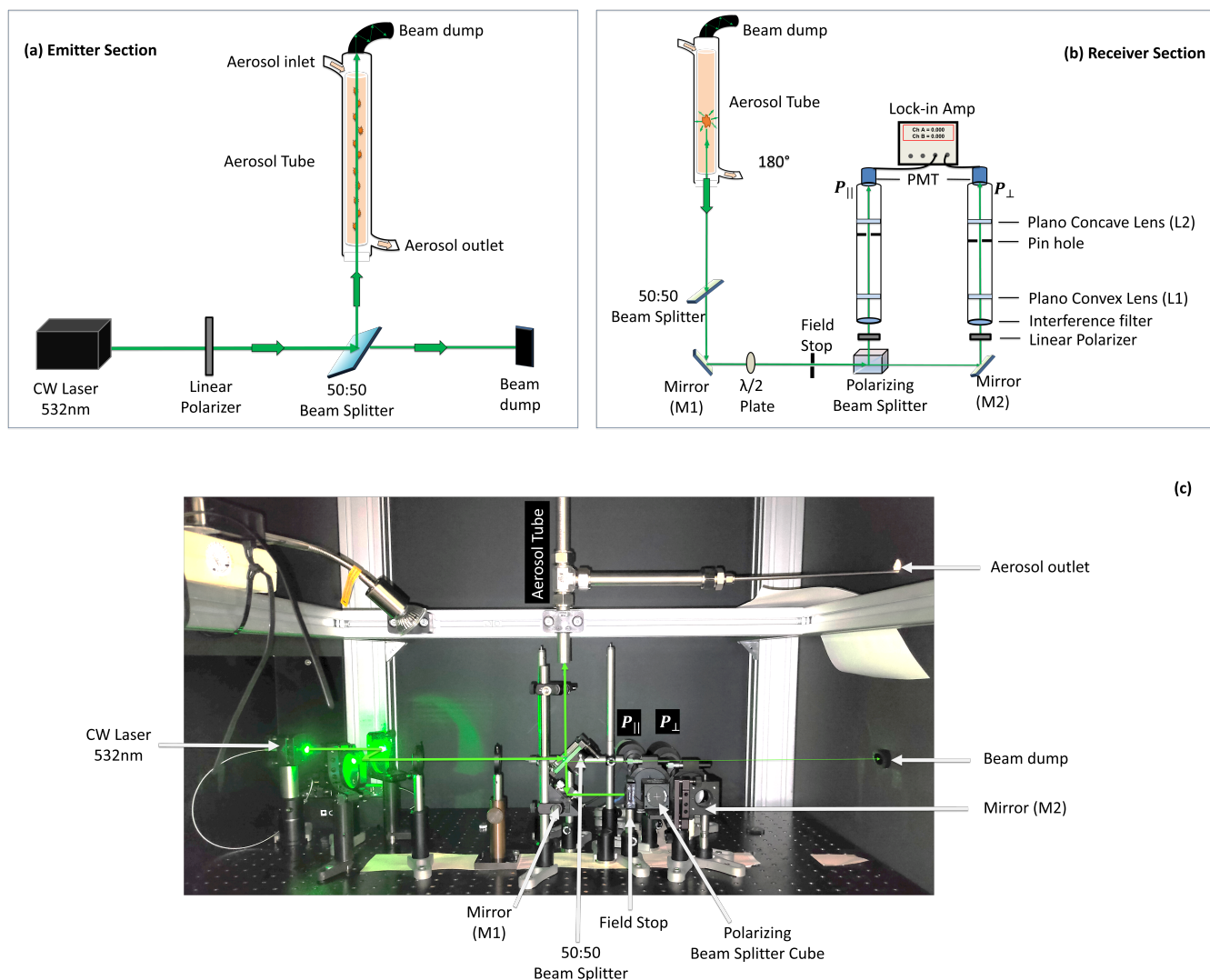


Figure 3. The optical setup of OLALA: (a) simplified block diagram illustrating the optical path of the emitted laser beam inside the aerosol tube following reflection from the 50:50 beam splitter, (b) block diagram illustrating the optical path of backscattered light from the aerosol tube to the co- and cross-polarized detection channels, (c) shows the optical setup in laboratory



1. Particle modulation: To suppress the unwanted background signals originating from the beam dump and reflections from the optical surfaces along the beam path, which were typically two to three orders of magnitude stronger than the particle backscattering signal, the particle concentration was modulated while the laser operated in continuous-wave mode. This method ensures that only the particle backscattering signal is periodically modulated, whereas the optical background remains constant. A lock-in amplifier was used for phase-sensitive detection, allowing only the signal component oscillating at the reference frequency to be extracted while effectively rejecting the background noise. The internal oscillator of the lock-in amplifier was used to generate a reference signal at a frequency of 0.05 Hz. The TTL (Transistor – Transistor Logic) output of this reference signal was used to control a pneumatic valve, hereafter referred to as the switch. This switch was used to alternate between air that was either particle-free or particle-loaded (OFF and ON states, respectively). The switch is controlled by the reference signal from the lock-in amplifier, switching between ON and OFF states every 10 seconds. When the switch is in the ON position, the aerosol is directed into the tube, resulting in an increase in particle concentration. In contrast, when the switch is in the OFF position, the particle-loaded air stream is redirected through a filter before entering the tube, leading to a decrease in particle concentration. This leads to a stable oscillation between a minimum and a maximum particle concentration. Figure 4 illustrates the temporal variation of the measured concentration of particles within the aerosol tube corresponding to the reference frequency of 0.05 Hz.

2. Working: Since the frequency of modulation of the particle flow is the same as the reference frequency of the lock-in amplifier, the lock-in amplifier extracts the desired backscattered signal of the particles from the noisy background. Because of the low reference frequency, a relatively long time constant of 100 s (or 300 s in some experiments) is utilized to average over multiple ON/OFF cycles and obtain stable, reliable measurements. To ensure sufficient data points, the measurements were recorded over an approximate duration of 2 to 3 hours. After a measurement with particles, we do a calibration with a half-wave plate. The calibration process takes another 3 to 4 hours. The process is illustrated in Fig. 5. A relatively low modulation frequency of 0.05 Hz was chosen because the particle concentration inside the aerosol tube does not respond instantaneously to the switching process. Sufficient time is required for the particles to travel through the 1 m long aerosol tube and for the concentration inside the aerosol tube to change after each ON/OFF transition of the switch. Using a higher modulation frequency would not allow the particle concentration to develop between switching cycles, thereby reducing the modulation contrast and weakening the extracted lock-in signal. The chosen frequency of 0.05 Hz provided adequate time for sufficient particle concentration changes while maintaining synchronized signal detection.

Due to the low operating frequency used in the particle modulation technique, the total duration of the experiment is relatively long, extending to approximately 7 hours (including the calibration time) for each size of a single particle type. However, this technique also eliminates the need for a separate background measurement. The lock-in amplifier measures the difference between the high backscattered signal and the low backscattered signal corresponding to the periodic variation in particle concentration at the modulation frequency of 0.05 Hz. During a particle-free measurement, the particle concentration does not change with the reference frequency but remains constant. Thus, no modulation of

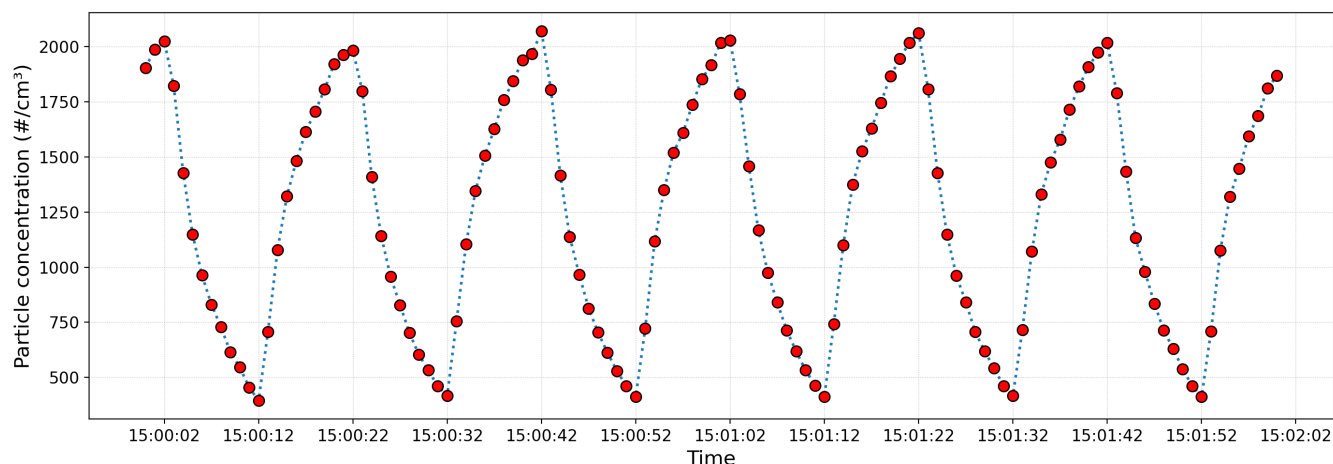


Figure 4. Temporal variation in particle concentration for 250 nm ammonium sulfate measured at the outlet of the aerosol tube. The plot shows the increase and decrease in particle concentration inside the aerosol tube when the switch turns between the ON/OFF positions every 10 seconds (switching frequency of 0.05 Hz).

the backscattered signal would occur at the reference frequency, and consequently, the lock-in amplifier would not detect any signal.

3. Calibration: After 3 hours of measurements with particles, we introduce a half-wave plate in the receiver section for calibration. The half-wave plate is placed between the dielectric mirror (M1) and the field stop (Fig. 3 b). The calibration procedure begins with measurements at the zero-degree position, corresponding to the orientation where the fast axis of the half-wave plate is parallel to the polarization direction of the incident beam. Data were recorded at this position for 30 minutes. Subsequently, the half-wave plate is rotated to $+22.5^\circ$ and -22.5° relative to the zero position. These orientations modify the polarization state of the backscattered light to $\pm 45^\circ$, respectively ($\Delta 90^\circ$ calibration, Freudenthaler, 2016). Measurements were performed for approximately 1.5 hours at $+22.5^\circ$ and -22.5° angular positions.

Figure 5 presents the temporal variation of the backscattered signal in the co- and cross-polarized detection channels during the measurement and calibration sequence for 250 nm ammonium sulfate and 450 nm ATD .

2.6 Data analysis and uncertainty estimate

Figure 5 shows an example of the backscattered signal measured in two channels for ammonium sulfate and ATD particles. The same measurement protocol was applied in all experiments. Owing to the long time constant, some time is needed to get the lock-in amplifier stabilized. Therefore, the initial 20–30 minutes of data (indicated by the shaded region in Fig. 5) are excluded from the statistical analysis. The statistical analysis was performed on a time-selected subset of the dataset.

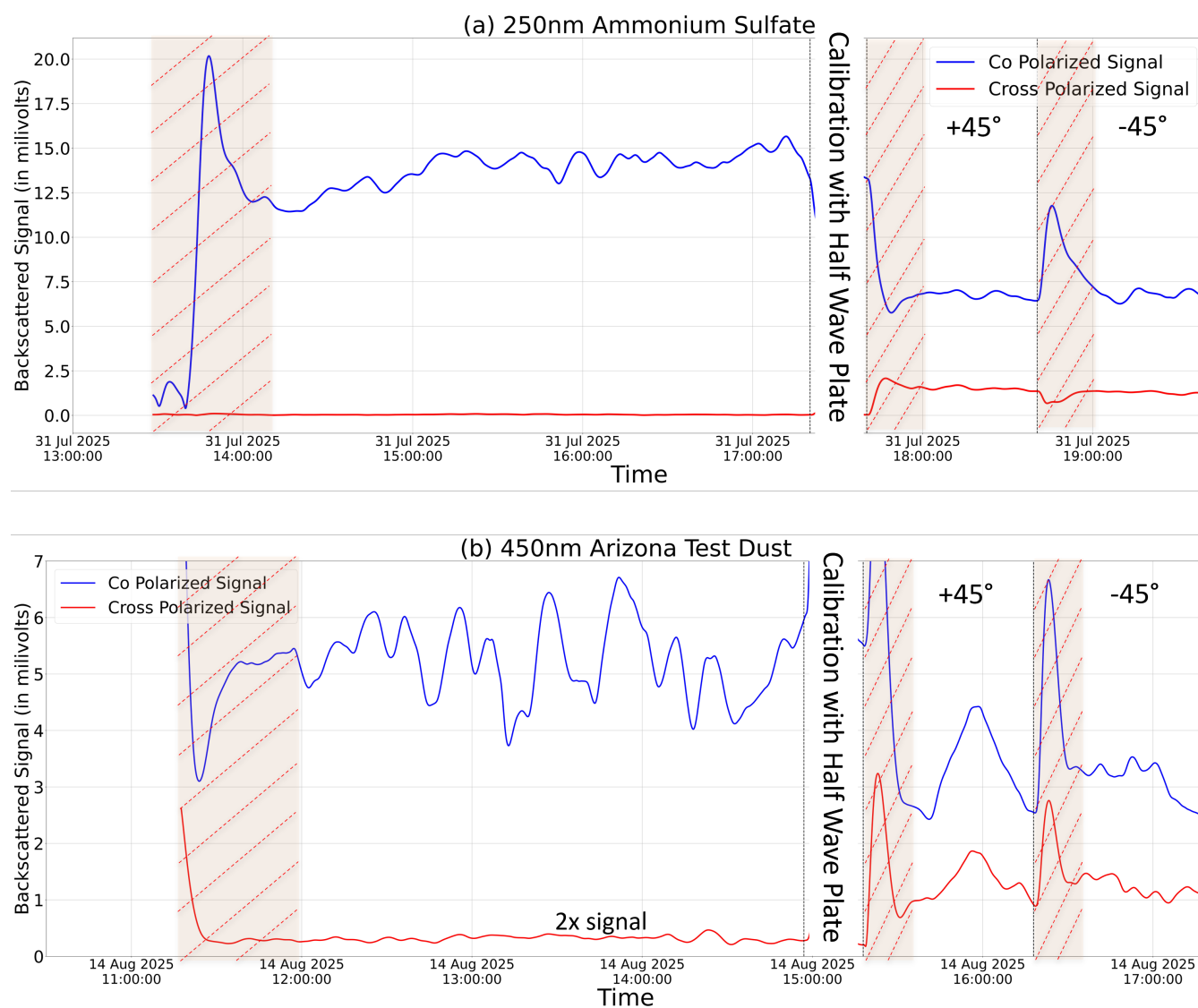


Figure 5. Temporal variation in backscattered signal for 250 nm ammonium sulphate (a) and 450 nm Arizona Test Dust (b), in the co-polar (blue) and cross-polar (red) channel. For 450 nm Arizona Test Dust, cross-polar signal is multiplied by a factor of 2 for better visibility. Due to a longer time constant, the lock-in amplifier takes some time to stabilize the detected signal. So, the first 20–30 minutes of measurements, shown by the shaded region in the plot, are not taken into consideration for data analysis.



The co-polarized signal ($I_{\parallel}$) and cross-polarized signal ($I_{\perp}$) were used to compute the signal ratio:

$$R = \frac{I_{\perp}}{I_{\parallel}} \quad (1)$$

260 To correct for differences in detector gain, a calibration constant C was determined from measurements of the signal ratio at $\pm 45^\circ$ polarization angles,

$$C = \sqrt{R_{+45^\circ} \cdot R_{-45^\circ}}, \quad (2)$$

and the depolarization ratio was then obtained as

$$\delta = \frac{R}{C} \quad (3)$$

265 as for a cleaned analyzer according to Freudenthaler (2016).

Extreme outliers influenced the signal mean. In order to remove their influence the following method was used: An iterative sigma clipping procedure was applied to each measured quantity, i.e., co-polarized signal and cross-polarized signal and also to the signal ratio R . In each iteration, we computed the sample mean (μ) and standard deviation (σ) for each quantity and rejected data points that were outside of a symmetric interval of $(\mu - 2\sigma) \leq x_i \leq (\mu + 2\sigma)$. This procedure was applied si-

270 multaneously to all three variables, so that a point was removed whenever it was inconsistent in any of these variables. After each clipping, the mean and standard deviation were recomputed on the remaining data, and the process was repeated (iterative sigma clipping) to mitigate the influence of extreme outliers on the estimated mean and standard deviation.

Because the measurements are acquired as time series with a finite instrumental time constant of 100 s, successive samples recorded at a 1 s interval are temporally correlated and therefore not statistically independent. To account for this correlation,

275 the effective number of independent samples was conservatively estimated by assuming approximately one statistically independent measurement per time constant, yielding $N_{\text{eff}} = \frac{N}{100}$. The standard error of mean was then calculated as: $\text{SEM} = \frac{\sigma}{\sqrt{N_{\text{eff}}}}$. The iteration was stopped when one of two objective criteria was met: (i) the effective number of samples fell below a pre-defined minimum, preventing excessive depletion of the dataset, or (ii) the relative change in the standard error of the mean (SEM) between successive iterations dropped below a threshold of 5 % for all variables, indicating convergence of the esti-

280 mated uncertainties. This ensures that only a small fraction of clearly inconsistent points (e.g. electronic spikes or transient mechanical disturbances) are removed, while the statistical properties of the signal are preserved.

Uncertainties in ratios of measured quantities were estimated using first order error propagation, under the assumption that the underlying measurements are uncorrelated.

3 Results and discussion

285 In this study, the linear depolarization ratio in the exact backscattering direction for fine mode aerosol particles at 532 nm wavelength under dry conditions ($\text{RH} < 10\%$) was measured. The particle size here refers to the different size fractions (A, B, C, D) selected by the DMA. Parameters characterizing each size fraction are given in Table 1, and the mean depolarization



ratio of all four size fractions for each aerosol type is presented in Table 2 and Figure 6. Concerning the four different size fractions, we will refer to them as sample A, sample B and so on in the following. The laboratory findings are discussed per aerosol type (Sect. 3.1) and then the cubical NaCl particles, which represent the only well-defined non-spherical particle shape, are compared against modelling results (Sect. 3.2).

Table 2. Particle linear depolarization ratio of different aerosol samples studied in the laboratory, measured at a wavelength of 532 nm for four different DMA-selected size fractions (A, B, C, D, see Table 1). The uncertainty depicts the mean measurement error.

Aerosol sample	Linear depolarization ratio			
	A	B	C	D
Ammonium Sulfate	0.026 ± 0.012	0.029 ± 0.012	0.076 ± 0.029	0.122 ± 0.040
Sodium Chloride	0.048 ± 0.017	0.045 ± 0.014	0.102 ± 0.035	0.167 ± 0.063
Arizona Test Dust	0.080 ± 0.018	0.131 ± 0.019	0.126 ± 0.013	0.168 ± 0.048
Saxonian Soil Dust	0.078 ± 0.027	0.117 ± 0.051	0.127 ± 0.048	0.152 ± 0.074

3.1 Laboratory results

3.1.1 Polystyrene Latex Particles

For spherical particles, Polystyrene Latex Particles (PSL) were used. The observed mean value of the depolarization ratio was 0.017 with a standard deviation of 0.002 for PSL particles of diameter of 1 μm . This result agrees very well with the theoretical predictions of Mie theory for spherical particles, which yields a depolarization ratio of 0. The low depolarization ratio measured in this study for these spherical PSL particles indicates that the experimental setup is correctly capturing the polarization response for low (or zero) depolarization ratios.

3.1.2 Ammonium sulfate

The depolarization ratio of ammonium sulfate was measured for four size fractions, as shown in Table 2. The low depolarization ratios observed for samples A and B (median diameter < 400 nm) of ammonium sulfate particles at a wavelength of 532 nm suggest that the particles are spherical, leading to weak depolarization of the backscattered light. The slightly higher depolarization ratios observed for samples C and D (median diameter > 600 nm) indicate increasing deviations from the spherical morphology, such as faceted crystalline shapes or particle aggregation. Electron micrographs of the ammonium sulfate particles presented in Fig. 8(a) of Sakai et al. (2010) show that some particles can exhibit irregular shapes due to the formation of aggregates of spheres. Figure 3(b) in Ueda (2021) shows electron micrographs of ammonium sulfate particles collected through a diffusion dryer. These images show that ammonium sulfate particles, produced from an aqueous solution and subsequently dried, predominantly occur as Type 1 (ball-type) and Type 3 (cluster-type) morphologies defined by Ueda (2021). The increase in depolarization ratio with particle size shown in Fig. 6 (a) may therefore be consistent with enhanced sensitivity

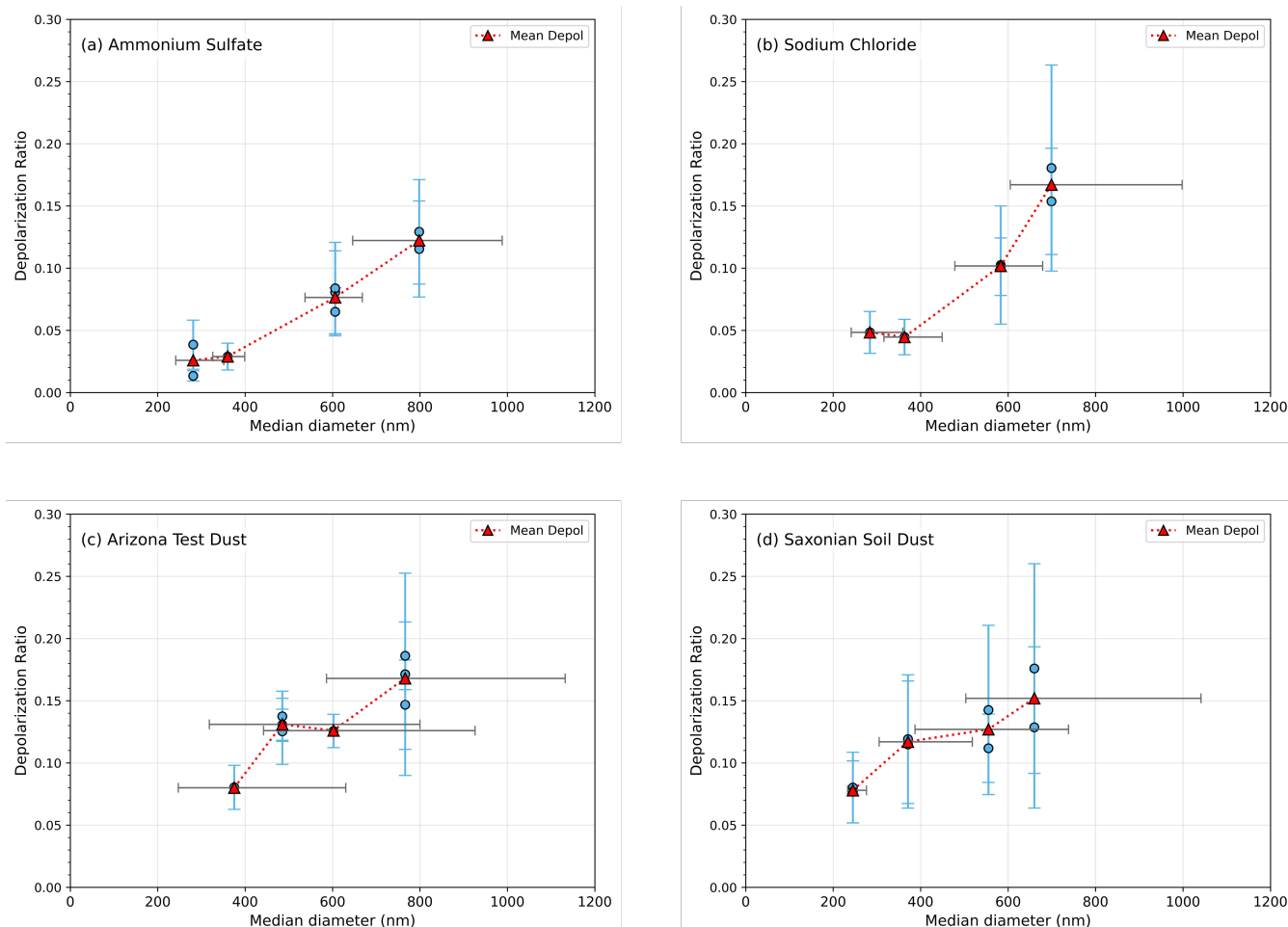


Figure 6. Variation in the linear depolarization ratio of different aerosol samples as a function of median optical diameter derived from the number size distributions measured with the POPS. (a) Ammonium sulfate, (b) Sodium chloride, (c) Arizona test dust, (d) Saxonian soil dust. The blue colored circular markers show the depolarization ratios measured in individual experiments. The red triangular markers indicate the mean depolarization ratio (values are provided in Table 2 as well) for each DMA-selected size fraction. The horizontal gray error bars represent the inter-quartile range (75th – 25th percentile) of the particle size distribution given in Tab. 1.



310 of backscattering polarization to particle non-sphericity at larger sizes. Laboratory experiments by Sassen et al. (1989) showed that small ammonium sulfate particles produced by evaporation of sulfuric acid droplets had depolarization ratios of about 0.02, while higher values of 0.10–0.15 were observed when the droplets partially crystallized after exposure to ammonia. Similarly, Sakai et al. (2010) reported a mean depolarization ratio of 0.04 ± 0.003 for crystalline ammonium sulfate with a mode diameter of 60 nm and a standard deviation of 2.4. In the same study, for ammonium sulfate droplets with a mode diameter of 80 nm and
315 a standard deviation of 2.3, a depolarization ratio of 0.01 with a standard deviation of less than 0.001 was measured.

3.1.3 Sodium chloride (NaCl)

Figure 6 (b) shows a general increase in the depolarization ratio of dry sodium chloride particles with increasing particle size. For sodium chloride, the depolarization ratios for samples A and B are low and very similar (agreement within measurement uncertainty), with an even slightly lower value for sample B. Median diameters for these two samples are 284 and 363 nm,
320 respectively. The similarity in depolarization ratios of these two samples can be attributed to the broadness of the generated size distributions. For samples C and D, depolarization values increased to 0.10 ± 0.03 and 0.17 ± 0.06 . We attribute these values to the non-sphericity of NaCl particles.

David et al. (2013) measured a lidar depolarization ratio of 0.04 ± 0.001 at a wavelength of 355 nm in exact backscattering direction for an ensemble of NaCl particles having size in the range of few hundred nanometers. Laboratory studies by Sakai
325 et al. (2010) found that for a sample of NaCl crystals with a mode diameter of 240 nm and a standard deviation of 1.9, the mean depolarization ratio was 0.21 ± 0.02 . Järvinen et al. (2016) measured a median depolarization ratio of 0.25 for dry NaCl. While these values are larger than ours, our laboratory measurements show good agreement with the theoretical calculations based on the Advanced Discrete Dipole Approximation presented in Sec. 3.2.

3.1.4 Arizona Test Dust (ATD)

330 The increasing trend in depolarization ratio with particle size for ATD is shown in Fig. 6 (c). Again, although the median diameter increased systematically for each DMA-selected size fraction, the POPS measurements revealed broad size distributions for all size fractions, with substantial overlap between adjacent populations. In particular, the distributions corresponding to samples B and C exhibited similar widths and overlapping inter-quartile ranges, which may have contributed to the comparable depolarization ratios observed for samples B and C.

335 Near-backscattering measurements by Järvinen et al. (2016) indicate depolarization ratios between 0.07 and 0.12 for ATD particles with a volume median diameter of approximately 500 nm at a backscatter angle of 178° . At a wavelength of 532 nm in the exact backscattering direction, Miffre et al. (2016) reported depolarization ratios of 0.35 ± 0.015 for an ATD sample containing particles with a diameter smaller than 400 nm and 0.30 ± 0.015 for samples containing particles up to 800 nm. More recently, Miffre et al. (2023) reported a depolarization ratio of 0.32 ± 0.01 for a finer ATD size distribution (particle diameter
340 < 800 nm). The depolarization ratios reported by Miffre et al. (2016) and Miffre et al. (2023) are higher than those observed in this study, but the origin of this discrepancy cannot be conclusively determined from the available data.



3.1.5 Saxonian Soil Dust

For the natural soil dust sample collected in Saxony, Germany, the depolarization ratio showed a similar increasing trend with particle size, as shown in Fig. 6 (d). For German soil dust collected in Brandenburg and Karlsruhe, depolarization ratios in the range of 0.12 to 0.25 were reported for particles with a volume median diameter between 300 nm and 800 nm, as shown in Fig. 7 by Järvinen et al. (2016). Sakai et al. (2010) observed depolarization ratios of 0.14 ± 0.03 for fine-mode Saharan dust and 0.17 ± 0.02 for fine-mode Asian dust at 532 nm. The samples observed by Sakai et al. (2010) were dominated by submicrometer particles and had a mode diameter of less than 200 nm and a standard deviation of 2.5. Miffre et al. (2023) reported a depolarization ratio of 0.23 ± 0.007 for Asian dust with a finer size distribution (diameter < 800 nm) at a wavelength of 532 nm. All of these values roughly agree with our results.

3.2 Comparison to model calculations for cubical particles

In this section, we compare the measured depolarization ratios of sodium chloride particles against modeled values. For this comparison, we used two different shape models to describe the morphology of sodium chloride: cubical particles and irregular hexahedra (TAMUdust2020, Saito et al., 2021). The ensemble sphericity of the irregular hexahedra was set to its maximum value $\Psi = 0.785$. The particle sphericity for a perfect cube is slightly higher at $\Psi \approx 0.806$. The optical properties of the cubical particles were calculated using the Advanced Discrete Dipole Approximation (ADDA version 1.4.0, Yurkin and Hoekstra, 2011) under random orientations and default settings.

For optical calculations, a conversion of the optical diameters D_{opt} , as reported by the POPS instrument, to the volume-equivalent diameters D_v , is necessary. For this purpose, two calibration curves of the POPS were calculated using the instrument specification: (1) The reference curve using Mie theory and the refractive index of PSL. (2) The correction curve using the TAMUdust2020 database and the refractive index of NaCl (at $\lambda = 405$ nm) which is $m_{\text{NaCl}} = 1.5663 + 0.0i$ (Eldridge and Palik, 1985). Due to the significant computational demand of running ADDA for large particle size parameters, an additional correction curve for the cubical particles is not considered due to insufficient computational resources. Instead, only the irregular hexahedra correction curve is employed to obtain the conversion.

The retrieved correction factor $\chi = D_v/D_{\text{opt}}$ is shown in Fig. 7. Since this correction factor is size-dependent, the particle number size distribution has to be corrected accordingly:

$$\frac{dN}{d\log D_v} = \frac{dN}{d\log D_{\text{opt}}} \frac{d\log(D_{\text{opt}})}{d\log(D_v)} = \frac{dN}{d\log D_{\text{opt}}} \chi \left(\frac{d\chi}{dD_{\text{opt}}} D_{\text{opt}} + \chi \right)^{-1}. \quad (4)$$

The bulk depolarization ratio is computed using the converted sizes and size distributions according to

$$\delta = \frac{b_{11} - b_{22}}{b_{11} + b_{22}}, \quad b_{ij} = \int_{-\infty}^{\infty} P_{ij}(180^\circ, D_v) C_{\text{sca}}(D_v) \frac{dN}{d\log D_v} d\log D_v, \quad (5)$$

with the scattering cross section C_{sca} .

Due to the aforementioned limited computational resources, the maximum cubic particle diameter for calculating the depolarization ratio is $D_v = 621$ nm, which implies that the upper 2% of the particle number size distribution is cut off for the cube

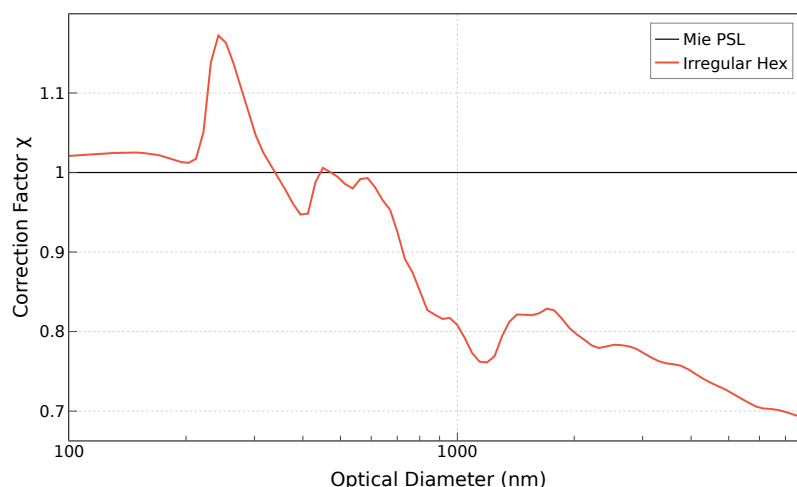


Figure 7. Size-dependence of the shape and refractive index correction for the POPS instrument, assuming an irregular hexahedral ensemble model ($\Psi = 0.785$).

calculations. In terms of the area size distribution, this corresponds to an upper cut off of about 9%.

Numerical results for the bulk depolarization ratio are shown in Fig. 8. The cubical shape model produces values that are
 375 consistently too low. The irregular hexahedra ensemble provides a good agreement for measurements A and B, but produces
 too large values for larger median diameters.

There are multiple factors that have to be considered in interpreting the numerical results. First, the cutoff of the size distribution for the cubical particles results in lower values as the depolarization ratio tends to increase with particle size. Second, the correction assumed an irregular hexahedral ensemble, also for the cubical particles. This potentially adds a systematic bias,
 380 which should be explored in future studies. Third, the lower boundary of the imaginary part of the refractive index in the TAMUdust2020 database is $n_k = 0.0001$, which results in a lower scattering signal compared to the non-absorbing case. In turn, especially for larger size parameters, this leads to a larger correction factor, also resulting in higher values of the depolarization ratio. While neither the cubical particles nor the irregular hexahedra ensemble can reproduce the measurements fully, they can be interpreted to describe lower and upper bounds on the depolarization ratio. Hence, they may show a measure of the
 385 sphericity of the laboratory-generated sodium chloride particle. This creates the hypothesis that the laboratory particles are not perfectly cubical, but rather exhibit an almost-cubical morphology.

4 Conclusions

In this study, we presented a new laboratory-based experimental setup that was developed to investigate the depolarization ratio of size-resolved aerosol particles in the exact backscattering direction ($180 \pm 0.2^\circ$). The presented setup provides a robust
 390 platform for investigating the scattering properties of irregularly shaped aerosol particles under well-defined conditions. It is

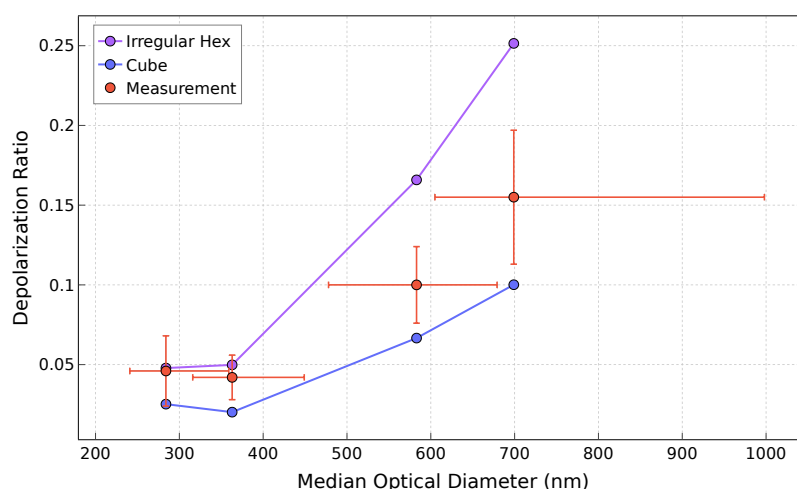


Figure 8. Comparison of measured versus modeled depolarization ratios of sodium chloride

therefore well suited for future studies aimed at improving the interpretation of polarization-sensitive lidar measurements and the modeling of aerosol optical properties.

The performance of the setup was evaluated using different aerosol types, including PSL, ammonium sulfate, sodium chloride, Arizona Test Dust, and Saxonian soil dust. PSL particles along with small ammonium sulfate particles exhibited the lowest depolarization ratio of 0.017 ± 0.002 and 0.026 ± 0.012 , respectively, among the investigated aerosols. This demonstrates that the setup accurately reproduces the negligible depolarization expected for spherical particles. While the small ammonium sulfate particles exhibited negligible depolarization, the depolarization ratio increased with increasing particle size, demonstrating the capability of the system to detect subtle changes in the morphology of predominantly spherical particles. For sodium chloride particles, which exhibit a cubic morphology with rounded edges and well-defined geometric properties, the laboratory measurements show good agreement with the scattering calculations. This further supports the capability of the setup to characterize non-spherical particles and provides a basis for comparison with light scattering calculations. Both dust samples showed substantially higher depolarization ratios, which is consistent with the irregular shape and surface roughness of mineral dust particles. Although theoretical modeling of irregularly shaped particles remains challenging, the developed setup provides reliable laboratory measurements that can be used to constrain and validate scattering models for mineral dust.

The measurements demonstrate that the system is capable of resolving the expected differences in depolarization behavior between spherical and non-spherical particles, as well as capturing the size dependence of the depolarization ratio for non-spherical aerosol particles. The measured depolarization ratios were generally consistent with values reported in the literature, although some differences remain. These discrepancies may arise from variations in experimental conditions, measurement geometry, as well as from differences or ambiguities in the particle size definitions used across the studies.

Future work will focus on extending the capabilities of the developed setup by implementing laser wavelengths 1064 nm and 355 nm to facilitate direct comparison with atmospheric lidar observations. Furthermore, we also plan to extend the measur-



able particle size range to include coarse-mode particles, which will enable more representative investigations of atmospheric dust.

415 *Data availability.* The particle size distributions and the individual particle linear depolarization ratios will be made available via zenodo upon final publication.

Author contributions. The paper was conceptualized by MoHa and ES. ES wrote the initial draft of the manuscript which was reviewed and edited by MoHa, DA, HW, MaHa, RE, and TO. ES, DA, MoHa and RE designed and developed the optical setup. MaHa and HW developed the microphysical setup. ES performed the experiments and data analysis with the support of MaHa and HW. TO provided the ADDA calculations. MoHa supervised the project.

420 *Competing interests.* The contact author has declared that none of the authors has any competing interests.

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