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Laboratory investigations of mineral dust near-backscattering depolarization ratios

E. Järvinen [a,*], O. Kemppinen [b], T. Nousiainen [b], T. Kociok [a], O. Möhler [a], T. Leisner [a], M. Schnaiter [a]

[a] Karlsruhe Institute of Technology, Institute of Meteorology and Climate Research, P.O. Box 3640, 76021 Karlsruhe, Germany
[b] Earth Observation, Finnish Meteorological Institute, P.O. Box 503, 00101 Helsinki, Finland

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ABSTRACT

Recently, there has been increasing interest to derive the fractions of fine- and coarse-mode dust particles from polarization lidar measurements. For this, assumptions of the backscattering properties of the complex dust particles have to be made either by using empirical data or particle models. Laboratory measurements of dust backscattering properties are important to validate the assumptions made in the lidar retrievals and to estimate their uncertainties. Here, we present laboratory measurements of linear and circular near-backscattering (178°) depolarization ratios of over 200 dust samples measured at 488 and 552 nm wavelengths. The measured linear depolarization ratios ranged from 0.03 to 0.36 and were strongly dependent on the particle size. The strongest size-dependence was observed for fine-mode particles as their depolarization ratios increased almost linearly with particle median diameter from 0.03 to 0.3, whereas the coarse-mode particle depolarization values stayed rather constant with a mean linear depolarization ratio of 0.27. The depolarization ratios were found to be insensitive to the dust source region or thin coating of the particles or to changes in relative humidity. We compared the measurements with results of three different scattering models. With certain assumptions for model particle shape, all the models were capable of correctly describing the size-dependence of the measured dust particle, albeit the model particles significantly differed in composition, shape and degree of complexity. Our results show potential for distinguishing the dust fine- and coarse-mode distributions based on their depolarization properties and, thus, can serve the lidar community as an empirical reference.

1. Introduction

Irregular mineral dust particles are ubiquitous in the Earth's atmosphere (e.g. [78,2,66]) and can affect Earth's climate by scattering and absorbing electromagnetic radiation (e.g. [82]). Therefore, it is important to be able to assess the radiative impact of dust correctly. For this, the information on the single-scattering properties of dust particles as well as their global coverage are needed. The global coverage of dust is retrieved using remote sensing methods that, also, rely on the accurate description of the mineral dust particle radiative properties. To accurately represent the dust scattering properties both in climate models and in remote sensing retrievals, we need to understand how these irregular particles interact with light. For this purpose laboratory studies on the optical properties of mineral dust particles have proven to be useful (e.g. [38,58,59,85]). Among the optical properties that can be measured in the laboratory, the backscattering

* Corresponding author.
E-mail address: emma.jaervinen@kit.edu (E. Järvinen).

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linear depolarization ratio is of particular importance for the interpretation of polarization lidar observations [68].

The polarization lidar technique is commonly used to study the vertical profiles of both tropospheric and stratospheric clouds [70] and aerosol distributions (e.g. [3,4]). A key measurement quantity in this technique is the particle backscattering linear depolarization ratio, δ_p, that describes the ratio of cross-polarized to polarized backscattering intensity. For a spherical isotropic particle, this ratio is zero [46], but for an irregular particle the ratio depends on the size, shape and refractive index of the particle [49]. Therefore, the depolarization ratio is considered to be an indicator of the particle non-sphericity, and is used to distinguish between liquid, mixed-phase and ice clouds as well as between aerosol types.

The separation of aerosol types is based on the information of their backscattering depolarization properties. Coarse-mode dust particles are known to have a high linear depolarization ratio compared to fine-mode aerosol (biomass burning smoke, urban haze) and, therefore, they can be distinguished from fine-mode aerosol particles (e.g. [80,81,77,64,83,4,5,22,7,57]). Moreover, the information of the coarse-mode particle fractions can be used to estimate the atmospheric ice nuclei concentrations [44]. On the other hand, the Aerosol Robotic Network (AERONET) sun/sky photometer observations show that a significant fraction of dust-related optical depth is linked with fine-mode dust [43]. This has led to a need to develop new techniques to determine the fraction of fine-mode dust to coarse-mode dust (e.g. [43]).

Despite the developments in polarization lidar retrieval techniques, the interpretation of the measurements are still hampered by the lack of accurate information on the depolarization properties of fine- and coarse-mode dust particles and other aerosol particles. In polarization retrievals the optical properties of dust particles are based either on empirically gathered data (e.g. [4]) or on modelled single scattering properties of dust particles (e.g. [15,9]). However, due to the computational limitations, most of the scattering models are restricted to model particles with simple geometries and uniform refractive indices, even though real dust particles are complex, irregular and inhomogeneous [61]. For instance, the spheroidal particle model has been established in the polarization lidar community [15], although recent comparisons with the spheroidal model and laboratory measurements show that the spheroidal model can only limitedly predict the mineral dust scattering properties [62,45]. As the computation power is increasing, new, more realistic models using complex geometries or inhomogeneous particles (e.g. [28,19,36,39,31,32]) could significantly improve the representation of dust particle scattering properties in lidar retrievals. However, these models still involve significant approximations and, therefore, validation of them require laboratory measurements of well-characterized dust samples.

While the development of "realistic" scattering models is an on-going work, remote sensing data are interpreted using a parametrization based on laboratory data or empirically gathered data rather than using a simplified scattering model. Mishchenko et al. [51] developed a retrieval method for the Advanced Very High Resolution Radiometer (AVHRR) based on experimentally measured phase function for quartz aerosol [40]. However, as single mineralogical components are not necessarily representative for atmospheric dust particles [61], new methods based on optical properties of atmospheric dust particle have been developed: Ansmann et al. [4] introduced the polarization lidar photometer networking (POLIPHON) method to separate between coarse-mode dust and fine-mode particles based on empirically gathered optical aerosol properties by Groß et al. [20] and Burton et al. [6]. Later the method was further developed to separate between fine- and coarse-mode dust particles [43] based on the laboratory work by Sakai et al. [68]. Despite the agreement between the laboratory and atmospheric dust depolarization properties, the authors stated that further laboratory studies are needed to justify the basic assumptions made in the retrievals.

There are only few laboratory studies available studying the backscattering or near-backscattering depolarization properties of natural mineral dust particles. Muñoz et al. [59] measured the whole scattering matrix, including the depolarization ratio, of Saharan dust particles. The angular range they covered was from 4° to 174°, but the setup was later improved to cover angles up to 177° [58]. The measured dust sample contained a mixture of particles with sizes ranging from a few microns to several hundred microns, so that the light scattering properties were dominated by the larger dust particles. The first step towards size-segregated measurements was taken by Sakai et al. [68]. The authors were able to experimentally show that the dust near-backscattering (angular range from 178.8° to 179.6°) depolarization properties are size-dependent by using an impactor to remove coarse-mode dust particles from the dust sample. The authors found out that the fine-mode dominated dust sample showed a significantly lower depolarization ratio (around 15%) than the coarse-mode dominated dust sample (δ_p around 40%). This laboratory study since served as an important basis for lidar studies (e.g. [4,43,21]). In a more recent study, Miffre et al. [47] measured at the exact backscattering angle ($180 \pm 0.2°$) depolarization ratio of two laboratory produced Arizona Test Dust size distributions. The authors performed the measurements at two wavelengths (355 and 532 nm), corresponding to common wavelengths used in lidar applications.

Still, significant experimental and theoretical work is needed to evaluate the scattering properties of natural dust particles. The experimental work is usually limited to uncertainties in aerosol properties, such as composition and size distribution. In this study, we address the latter problem and present size-segregated laboratory measurements of near-backscattering linear and circular depolarization ratios for mineral dust particles of natural and industrial origin. Here, the term "near-backscattering" is referred to a scattering angle of 178°, whereas "exact backscattering" takes place at 180°. We combined an aerosol chamber with an aerosol dispersion technique using multiple impactors to produce well-controlled airborne and mono-modal aerosol populations with a narrow mode width. The aerosol size distribution was measured using a combination of an aerodynamic particle sizer