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UV–VIS depolarization from Arizona Test Dust particles at exact backscattering angle

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ABSTRACT

In this paper, a controlled laboratory experiment is performed to accurately evaluate the depolarization from mineral dust particles in the exact backward scattering direction ($\Theta = 180.0 \pm 0.2°$). The experiment is carried out at two wavelengths simultaneously ($\lambda = 355$ nm, $\lambda = 532$ nm), on a determined size and shape distribution of Arizona Test Dust (ATD) particles, used as a proxy for mineral dust particles. After validating the set-up on spherical water droplets, two determined ATD-particle size distributions, representative of mineral dust after long-range transport, are generated to accurately retrieve the UV–VIS depolarization from ATD-particles at exact backscattering angle, which is new. The measured depolarization reaches at most 37.5% at $\lambda = 355$ nm (35.5% at $\lambda = 532$ nm), and depends on the particle size distribution. Moreover, these laboratory findings agree with T-matrix numerical simulations, at least for a determined particle size distribution and at a determined wavelength, showing the ability of the spheroidal model to reproduce mineral dust particles in the exact backward scattering direction. However, the spectral dependence of the measured depolarization could not be reproduced with the spheroidal model, even for not evenly distributed aspect ratios. Hence, these laboratory findings can be used to evaluate the applicability of the spheroidal model in the backward scattering direction and moreover, to invert UV–VIS polarization lidar returns, which is useful for radiative transfer and climatology, in which mineral dust particles are strongly involved.

1. Introduction

With emission rates as large as 1000–3000 Tg yr^{-1} from the Earth's surface, mineral dust is one of the major contributors to the Earth's global aerosol load [1]. This abundant aerosol is uplifted into the atmosphere through favourable winds [2] and can be transported by advection over several thousands of kilometres, hence affecting the Earth's climate at both local, regional and global scales. The latest IPCC report [3] underscores the key role of mineral dust particles on the Earth's radiative budget, both directly, at a magnitude comparable to that of greenhouse gases, and indirectly, by acting as cloud condensation nuclei and promoting new particles formation events, as we have recently reported [4]. However, quantifying the impact of mineral dust on the Earth's climate still remains a challenging task, mainly because mineral dust particles present a wide range of sizes, shapes and chemical composition, which varies with time and space and are modified during advection by chemical reactivity [5], mixing with other aerosols, such as sea-salt particles, or settling by gravitation [6]. Hence, after long-range transport, dust particles may exhibit characteristic shapes that can be very different from those observed close to their source region [7] and the dust particles residence time in the Earth's atmosphere is sometimes increased from hours to weeks [8], which further reinforces the impact of mineral dust particles on the Earth's climate. Hence, after long-range

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transport, the microphysical properties of atmospheric dust particles are interesting to study for climatology, air quality and human health purposes.

Mineral dust particles contribute directly to the Earth's radiative budget through light scattering, absorption and emission [9]. Since most mineral dust particles are highly irregularly-shaped, no analytical solution of the Maxwell's equations exists for light-scattering by these particles, since they are difficult to represent mathematically in climate models. Hence, to increase the knowledge on desert dust particles, field experiments have been performed to measure the properties of desert dust layers in the atmosphere, either with ground-based [10–12] or satellite-based [13] polarization lidars. In this context, polarization lidars have been widely used [10–12] to discriminate non-spherical dust particle backscattering from spherical particle backscattering, as we have previously demonstrated by developing the scattering matrix formalism of a mixture, composed of spherical and non-spherical particles [14]. We then extended this methodology to the practically important case of three-component particle external mixtures, where non-spherical desert dust particles are mixed with cubic sea-salt particles and water-soluble species [15], by coupling a sensitive and accurate UV-VIS polarization lidar [11] with T-matrix numerical simulations [9]. However, to successfully retrieve the microphysical properties of mineral dust particles from such polarization lidar measurements, a robust inversion algorithm is required and for this, accurate input parameters are needed, at least to avoid confusions between mineral dust, sea-salt particles and other non-spherical particles. Hence, a precise knowledge of the dust particles depolarization is required [10–12,16,17]. To evaluate the dust particle depolarization, care should be taken when using polarization lidars [10–17], because the lidar-measured depolarization is that of a mixture of particles of different types, which is smaller than the dust particle depolarization, as we demonstrated [14] by developing the scattering matrix formalism of a mixture, composed of spherical and non-spherical particles. Formally, in the lidar backward scattering direction, the dust particle depolarization is determined by the ratio F_{22}/F_{11} from the scattering matrix elements F_{11} and F_{22}, which mainly depend on the size, the shape and the chemical composition of the dust samples [9]. Additional subtle effects such as inhomogeneity [18–20] or surface roughness [21] may also modify the particle depolarization. Hence, to accurately retrieve mineral dust microphysical properties, scattering matrix elements have to be precisely determined, either with numerical simulations (see http://www.scattport.org) or/and with controlled laboratory experiments (see http://www.astro.uva.nl/scatter), both methodologies being complementary.

Numerical simulations are indeed becoming realistic [19,22] with improved accuracies allowing an evaluation of the influence of the dust particles inhomogeneity [18–20] and surface roughness [21] on the scattering matrix elements. Such numerical simulations are now currently performed over the whole scattering angle range, hence including the exact backward scattering direction $\theta=\pi$. However, to be fully beneficial, these numerical simulations should be validated through controlled laboratory experiments, to help developing more accurate light-scattering models.

However, in the published literature, backscattering of light has been observed and applied only for dense media such as solid GaAs crystals or solid biological tissues. For aerosols, laboratory experiments have been carried out only close to the 180°-backward scattering direction [23–28] so that, as recently underscored [29], in laboratory experiments, "the phase matrix can be obtained only at specific wavelengths and in limited angular scattering regions, for example, from 3° to 177°". Hence, ratios of scattering matrix elements (i.e. normalized at 30° scattering angle) rather than absolute scattering matrix elements can be determined [30], which may limit the direct applicability of the measured scattering matrix elements for radiative transfer calculations [31]. To cover the exact backward scattering direction, polynomial extrapolations have been proposed and a so-called synthetic scattering matrix has been built [30–32]. However, as analysed by Liu [33] or in [28], the assumptions inherent to extrapolations need to be checked with controlled laboratory experiments, as recommended in [29]. According to Hovenier [32], laboratory scattering experiments should then be performed from scattering angles around 177° to the exact backscattering angle to address the slope of the scattering matrix elements that tends to zero in the backward direction.

Hence, a controlled laboratory experiment that would specifically address the π-scattering angle for mineral dust particles is highly desirable. Ideally, such a laboratory backscattering experiment should be conducted at several wavelengths, under the far-field single scattering approximation [34], to ease the comparison with numerical models [35,36]. In David et al. [37], we reported on the principle of a controlled laboratory experiment addressing light depolarization by aerosols in the exact backward scattering direction with accuracy ($\theta=180.0\pm0.2°$). This prototype could however only run for spherical particles such as water droplets or salt particles above the crystallization or deliquescence points. For such spherically symmetric particles, the Lorentz–Mie theory could however have been applied, although it was to our knowledge the first time that this theory was experimentally checked in the exact backscattering direction for particles in ambient air. Addressing mineral dust particles backscattering is a priori much more difficult since backscattering from non-spherical particles is generally weaker than for spherical particles [38].

The present study is precisely dedicated to the measurement of direct backscattering from dust particles and complements previous laboratory experimental set-ups operating close to the exact backward scattering direction [23–28]. Our concern is to develop a controlled laboratory experiment to address the issue of mineral dust particle backscattering after long-range transport. The novelty of this work is hence twofold. Firstly, by addressing mineral dust particle backscattering in a controlled laboratory experiment, we evaluated the particles depolarization at the specific π-angle and at two wavelengths, namely at $\lambda=355$ nm and $\lambda=532$ nm, at which most