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Interpreting the Particle Linear Depolarization Ratio spectral dependence of aged stratospheric smoke using T-matrix simulations

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In the summer of 2017 western Canada experienced record breaking wildfires accompanied by intense Pyro-cumulonimbus activity, which favored the injection of large amounts of smoke into the lower stratosphere. The smoke particles transported in the stratosphere and reached Europe after almost 10 days of transport. Lidar measurements from Leipzig (Germany) report an optically thick smoke plume measured at an altitude of 15 -16 km (Ansmann et al., 2018), originating from Canada. While smoke particles are not expected to significantly alter the polarization state of the incident radiation, the analysis of their optical properties over Leipzig revealed large values of the Particle Linear Depolarization Ratio (PLDR) and furthermore a strong spectral dependence with values of $\sim$ 22.4 %, $\sim$ 18.4 % and $\sim$ 4.3 % at 355, 532 and 1064 nm respectively (Haarig et al., 2018). Similar values of PLDR have been observed before via airborne measurements (Burton et al., 2015).
Smoke particles once emitted in the atmosphere, tend to form fractals of soot aggregates with complicated shapes, but as particles age they tend to become more hydrophilic and are usually found partially or even completely encapsulated in water-soluble materials e.g. sulfates (Ishimoto et al., 2018) forming near-spherical shapes. Simulations carried out by Bi et al. (2018), revealed that this near spherical shape of the particles, can produce large values of PLDR at one or several wavelengths. Furthermore, Mishchenko et al. (2018) showed that the PLDR values measured by Burton et al. (2015), can be reproduced for non-spherical smoke particles containing substantial amounts of weakly or even non-absorbing materials.
Based on the above, we present T-matrix simulations (Mishchenko et al., 1996) aiming to reproduce the measured depolarization ratio values above Leipzig considering near-spherical particles with different degree of sphericity, effective radii and refractive indices.
It is found that the near-spherical shape has the potential to reproduce successfully the large spectral dependence of PLDR. Specifically, for particles with effective radius r eff = 0.45 μm, axial ratio a/b = 1.15 and refractive index m = 1.45 + 0.04i, values of $\sim$ 21 %, $\sim$ 17 % and $\sim$ 2 % are found at 355, 532 and 1064 nm respectively. Simulations contacted for more complicated morphologies (Chebyshev particles) showed that the PLDR values of stratospheric smoke cannot be reproduced. Further investigation of the particular atmospheric processes associated with smoke particles aging that may lead to these observations is underway.