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Stratospheric aerosol particle size distribution based on multi-color polarization measurements of the twilight sky

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ABSTRACT

Polarization measurements of the twilight background with Wide-Angle Polarization Camera (WAPC) are used to detect the depolarization effect caused by stratospheric aerosol near 20 km altitude. Based on a number of observations in central Russia in spring and summer 2016, we found the parameters of lognormal size distribution of aerosol particles. This confirmed the previously published results of the colorimetric method as applied to the same twilights. The mean particle radius (about 0.1 μm) and size distribution are also in agreement with the recent data of *in situ* and space-based remote sensing of stratospheric aerosol. The methods considered here provide two independent techniques of the stratospheric aerosol study based on the twilight sky analysis.

1. Introduction

The history of stratospheric aerosol observations covers more than 50 years, after the first balloon measurements provided by Junge, Changnon, and Manson (1961). Aerosol concentration reaches the maximum near the altitude of 20 km (the Junge layer). Long-lasting series of balloon experiments started in the 1970s (Hofmann & Rosen, 1980) showed that stratospheric aerosol particles are droplets of sulfur acid solution (Rosen, 1971). Their number sufficiently increases after intensive volcanic eruptions due to the emission of SO_2 into the stratosphere. A combined analysis of balloon and lidar measurements in 1970s described in Swissler, Hamill, Osborn, Russell, and McCormick (1982) showed the effects of several volcanoes, of which the most noticeable ones were Fuego in 1974 and St. Helens in 1980. The eruption of El Chichon in 1982 significantly changed the aerosol characteristics for the following five years (Hofmann & Rosen, 1987). During these years, stratospheric aerosol was studied by balloon techniques (Hofmann & Rosen, 1982), lidars (McCormick, Swissler, Fuller, Hunt, & Osborn, 1984), and satellites (SAGE II, Thomason, Poole, & Deshler, 1997). The most recent major eruption, which was also the most powerful one in the 20th century, was Mt. Pinatubo in 1991, whose significant effect on stratospheric aerosol lasted for several years (Bauman, Russell, Geller, & Hamill, 2003; Deshler, Hervig, Hofmann, Rosen, & Liley, 2003; Jäger, 2005; McCormick, Thomason, & Trepte, 1995).

In the beginning of 21th century, the effects of several minor eruptions were recorded by space- based techniques (SCIAMACHY, von Savigny et al., 2015) and lidar remote sensing (Burlakov, Dolgii, & Nevzorov, 2011; Ridley et al., 2014). During the recent years, stratospheric aerosol remained in its background conditions, and it was the longest volcanically-quiet period since the beginning of regular stratosphere observations. However, its properties seem to change over time. A positive trend of background aerosol was noticed by Solomon et al. (2011). Earlier measurements by Hofmann and Rosen (1980) had shown an increase in the stratospheric aerosol amount compared to the data of Junge et al. (1961). However, this comparison could be affected by the low accuracy of

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Junge's results. The 30-year survey (Deshler et al., 2006) did not show any statistically significant trend of background stratospheric aerosol. This question is important, since this aerosol can have partially anthropogenic origin, related to urban emissions of carbonyl sulfide (Crutzen, 1976) and sulfur dioxide (Brock, Hamill, Wilson, Jonsson, & Chan, 1995).

One of the basic characteristics of stratospheric aerosol related to the physical processes of its formation that could be found by the observations is the size distribution of particles. Its determination was the fundamental goal of the balloon measurements (Deshler et al., 2003) and space-based analysis (Bingen, Fussen, & Vanhellemont, 2004). It was shown that under the background conditions, the size distribution is well described by a lognormal function:

$$dn(r) = \frac{N}{\sqrt{2\pi}\,\ln\sigma}\exp\left(-\frac{\ln^2(r/r_0)}{2\ln^2\sigma}\right)d\ln r, \tag{1}$$

where N is the total number of particles; r_0 is the mean radius; and $\sigma > 1.0$ is the distribution width. In volcanically-perturbed conditions, the distribution becomes bimodal with the second peak characterized by larger r_0 and related to particles of volcanic origin. The typical values of background lognormal distribution parameters found by Deshler et al. (2003) and by other measurements are $r_0 \sim 0.08$ μm, $\sigma \sim 1.6$, being practically independent of the time. However, the mean radius depends on the altitude, with a maximum near 20 km, in both background and post-volcanic conditions (Hofmann & Rosen, 1982).

At the present time, the stratospheric aerosol characteristics are being measured within the OSIRIS experiment onboard the Odin satellite (Bourassa, Rieger, Lloyd, & Degenstein, 2012) and OMPS LP experiment held onboard the NPP satellite (DeLand, 2017) using the limb viewing geometry. The light-scattering properties at different wavelengths can be compared to obtain the size distribution of aerosol particles. Bourassa, Degenstein, and Llewellyn (2008) found that the size distributions are in good agreement with the ones described above.

An analysis of light scattering by stratospheric aerosol for the size determination can be performed from the ground using the multi-wavelength lidar technique (Di Girolamo et al., 1995); in this case, backscattering (by angle 180°) is considered. It can be also done during twilight, when the stratosphere is illuminated by solar radiation while the troposphere is not. A twilight analysis can not have a good altitude resolution; however, it can involve a wide range of scattering angles, increasing the accuracy of the size distribution retrieval.

Ugolnikov and Maslov (2018) used the data on the twilight sky color (or intensity ratio at different wavelengths) to find the aerosol scattering component of the twilight background and to study the particle properties. Despite the approximate (or even empirical) nature of the method, the size distribution parameters were found to be close to the ones determined by the space-based or balloon experiments. In this paper, we use an independent approach to the same observational data and find the aerosol characteristics by the polarization analysis, comparing them with the color data. Twilight sky polarization is sensitive to aerosol scattering, and this fact helped to detect the aerosol from the Tavurvur (Rabaul) volcano in 2006 (Ugolnikov & Maslov, 2009), using just one-wavelength measurements near the zenith. The color and polarization analyses are found to be effective for noctilucent cloud study (Ugolnikov, Galkin, Pilgaev, & Roldugin, 2017; Ugolnikov, Maslov, Kozelov, & Dlugach, 2016). The background stratospheric aerosol analysis is more difficult due to less contribution to the total sky brightness; however, multi-wavelength measurements over a large part of the sky make it possible.

2. Observations and polarization effects of aerosol

Multi-color polarization measurements of the twilight sky background were conducted in Chepelevo (55.2°N, 37.5°E, 50 km southwards from Moscow) in March–July 2016. The observations were performed with Wide-Angle Polarization Camera (Ugolnikov & Maslov, 2013a, 2013b). The field diameter was 140°; the points under consideration were at zenith angles of up to 55–60°. RGB-color CCD Sony QHYCCD-8 was used; the effective wavelengths were equal to 461, 540, and 624 nm for wide RGB channels, respectively, FWHM of each band is about 90 nm. The R channel was corrected with an IR-blocking filter with the threshold at 680 nm. The exposure times varied from 3 ms to 30 s, depending on the twilight stage. Star images at different zenith angles on the night frames were analyzed to identify the camera position, flat field, and atmosphere transparency.

The procedure considered herein is quite similar to the one based on the same observations and described by Ugolnikov and Maslov (2018). However, we took polarization and its difference in R and B bands instead of the sky intensity and color. We use the polarization data in the solar vertical (the celestial major circle containing the zenith and the Sun); the point position is characterized by the zenith angle ζ, which is positive in the dusk area and negative in the opposite part of the sky. The solar zenith angle was denoted as z. The picture of light scattering geometry during the twilight is shown in Fig. 1.

When describing the polarization properties of the sky background in the solar vertical, it is sufficient to find the normalized second Stokes component:

$$q(\zeta, z) = -\frac{I_2(\zeta, z)}{I(\zeta, z)} = -p(\zeta, z)\cos 2A(\zeta, z). \tag{2}$$

Here, I and I_2 are the first and second Stokes components, p is the sky polarization degree, and A is the polarization direction angle with respect to the solar vertical (the sky major circle containing the Sun and zenith). During the light twilight (solar zenith angle less than 95–96°) the angle A is close to 0° or 90° in this vertical, so the quantity q is usually equal to $\pm p$ and can be simply called the polarization. High above the horizon, the value of q is positive due to the properties of Rayleigh scattering. It decreases and can be even negative closer to the horizon both in the dusk and opposite parts of the sky; the main reason is multiple scattering contribution