Stratospheric aerosol extinction profiles retrieved from twilight sky spectral measurements above Georgia, South Caucasus in 2021-2022.

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## Abstract

Narrow field-of view spectral measurements of twilight sky brightness as a function of solar zenith angle in the 89°-95° range allow to retrieve lower stratospheric and tropospheric aerosol extinction profiles. The measurements were carried out over Tbilisi, Georgia, South Caucasus during 2021-2022 in the 700-800 nm wavelength range using a SBIG ST9 CCD camera and a SBIG SGS spectrograph. The Monte Carlo code Siro, developed in the Finnish Meteorological Institute was used to design a forward model. Aerosol extinction profiles at 780 nm were retrieved using the Levenberg-Marquardt algorithm.

By analyzing the dependence of the color ratio (ratio of extinctions measured at different wavelengths at same solar zenith angle) on solar zenith angle, it is possible to detect changes in the size distribution of aerosol present in the atmosphere. The color ratio can provide complementary information to twilight sky brightness as a function of solar zenith angle, helping to create a more complete picture of the state of the atmosphere. We show it on example of Etna eruption cloud.

Stratospheric aerosol optical depths were retrieved and a patchy nature of the stratospheric layer was observed.



## The measurements (Fig.1)

Twilight sky brightnesses were measured from the ground at solar zenith angle range 89°-95° at wavelengths 780nm and 870 nm. SBIG ST-9XE CCD camera equipped with SBIG SGS spectrograph was used. The presence of a strong aerosol layer significantly disturbs the brightness of the twilight sky (Fig,2, left), causing both an increase and a decrease in brightness depending on the range of heights and thickness of the layer, the presence of a second layer above the first, and the level of transparency in the interval between the two layers.

Fig.2, Twilight sky brightness dependences on solar zenith angle (left) and corresponding color ratio dependences on solar zenith angle (right).



## The retrieval.

A spherical single scattering forward model with multiple scattering correction factors calculated using a Monte Carlo code (Oikarinen et al., 1999) was developed. To amplify the dynamics of the twilight curve and to remove all constant calibration factors, the measurement vector y was presented as the differences of logarithm of intensity vs. solar zenith angle:



An error-weighted least-squares fitting which aimed to retrieve the aerosol extinction profiles was performed by means of a Levenberg–Marquardt algorithm.

 $\chi^2 = [\mathbf{y} - f(\mathbf{x})]^T \mathbf{S}_{s}^{-1} [\mathbf{y} - f(\mathbf{x})] + (\mathbf{x} - \mathbf{x}_{a})^T \mathbf{S}_{a}^{-1} (\mathbf{x} - \mathbf{x}_{a}),$ 

# Aerosol signature on twilight curve.

Fig. 2 shows two sets of the twilight sky brightness curves and corresponding color ratios at different solar zenith angles (SZA). The left plot of Fig. 2 shows the dependence of twilight sky brightnesses on solar zenith angle, while the right plot shows the dependence of color ratios  $(\log_{10}(I_{780}nm/I_{870}nm))$  on solar zenith angle.

There are two groups of curves shown in Fig. 2: one set of curves measured during autumn 2019 when the stratospheric aerosol layer was disturbed after the Raikoke eruption, and another set of curves measured during summer 2022 when the stratospheric layer was not significantly disturbed in the Northern hemisphere.

Although wavelengths 780 and 870 nm are quite close one to another the color ratio curves form two groups attributed to post-volcanic and volcanically quiet period are quite distinct.

The difference in color ratio between the two data groups shown in the right plot of Fig. 2 is likely to be related to the particle size distribution in the stratospheric aerosol layer. Specifically, larger particles in the aerosol layer are expected to scatter more light at longer wavelengths, which could explain the difference in color ratio between the post-volcanic and volcanically quiet periods.

The low color ratio values at solar zenith angles less then 91° for the 2022 year data is likely to be connected with the presence of dust in the lower troposphere. This is because larger dust particles in the troposphere can scatter more light at longer wavelengths, which could contribute to the decrease in the color ratio as the solar zenith angle decreases. However, when the solar zenith angle exceeds 91°, the lower tropospheric dust layer is already in the Earth's shadow and does not contribute significantly to the twilight sky brightness. Therefore, the decrease in the color ratio at low solar zenith angles is likely to be attributed to the presence of lower tropospheric dust particles, and this effect becomes negligible at higher solar zenith angles.

## Etna eruption

A minor eruption occurred at Mount Etna on February 11, 2022, with an eruptive column that reached up to 10 km in height. The emitted sulfur dioxide (SO<sub>2</sub>) cloud passed over Georgia the following day, as shown in Fig. 3. While it may not have been enough time for sulfuric acid droplets to grow, small ash particles could have been transported with the cloud. Fig. 4, left plot, shows a small extinction enhancement at around 10 km altitude (red curve), which represents the extinction profile after the eruption, but the difference between the two extinction profiles is within the limits of uncertainties.

In this case, the color ratio could be helpful in identifying changes in the aerosol load of the atmosphere. The increase in the slope between 92°-93.5° on the red curve compared to the blue curve (Fig.4, right) indicates a change in the size distribution of the aerosols. This change in the color ratio suggests that there could be a presence of ash particles which are larger than those of stratospheric sulphur aerosol in the atmosphere after the eruption, which could have contributed to the observed extinction enhancement. Therefore, the color ratio can provide additional information to help identify changes in the aerosol content of the atmosphere.



Fig. 3. Etna eruption in Feb. 11, 2022. Blue area -SO<sub>2</sub> cloud emitted by Etna in Feb. 11, 2022 (left) and the same cloud in Feb.12,2022 (right), red point – Tbilisi, Georgia

(image source: Suomi NPP/OMPS, worldview.earthdata.nasa.gov)



## **Fig.4** Left-aerosol extinction profiles retrieved from twilight measurements before (in blue) and after (in red) a cloud from Etna passed through (dashed lines mark tropopause altitude). The right plot shows the corresponding color ratios.

Fig.5, Stratospheric aerosol optical depths, July-September 2022. Some of the measurements (red points) show significantly higher optical depths than the majority of

#### Stratospheric optical depth

**Fig. 5** shows the stratospheric aerosol optical depths obtained by integrating the extinction profiles retrieved from twilight measurements at 780 nm above the tropopause for the period of July-September 2022. Some of the measurements (red points) show significantly higher optical depths than the majority of the measurements (blue points).





If we exclude the high optical depth values (red points) and average the remaining data, the averaged optical depth is about 0.002. However, if we include the high optical depth values, the averaged optical depth can be as high as 0.01. In simpler terms, the measurements of stratospheric aerosol optical depth during the period July-September 2022 indicate that there was a low aerosol level throughout the period, but some measurements showed much higher aerosol levels than others. These high levels of aerosols were observed less frequently, but had much higher optical depths compared to the more commonly observed aerosol levels. Therefore, the aerosol level was not uniform and there were some areas where aerosol levels were significantly higher than others.