Stratospheric and upper tropospheric aerosol extinction profiles over Georgia, South Caucasus in 2009-2010
as retrieved from ground-based spectral measurements of twilight sky brightness.

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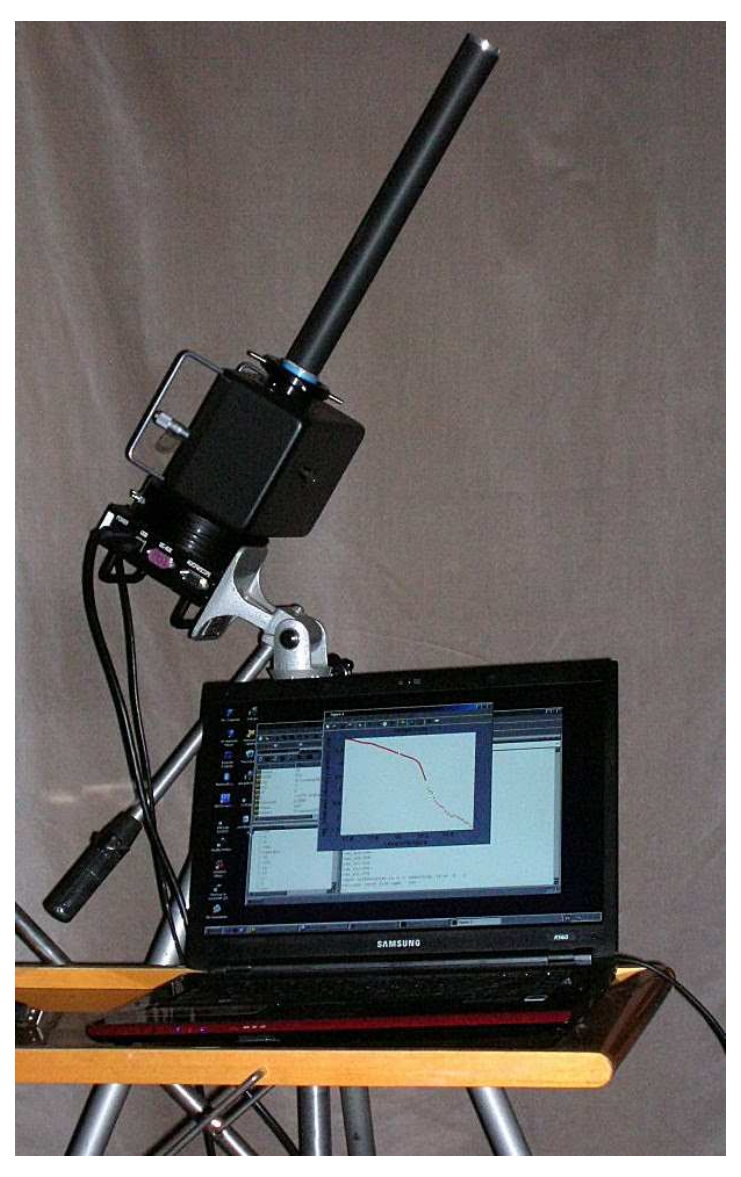
1.Introduction

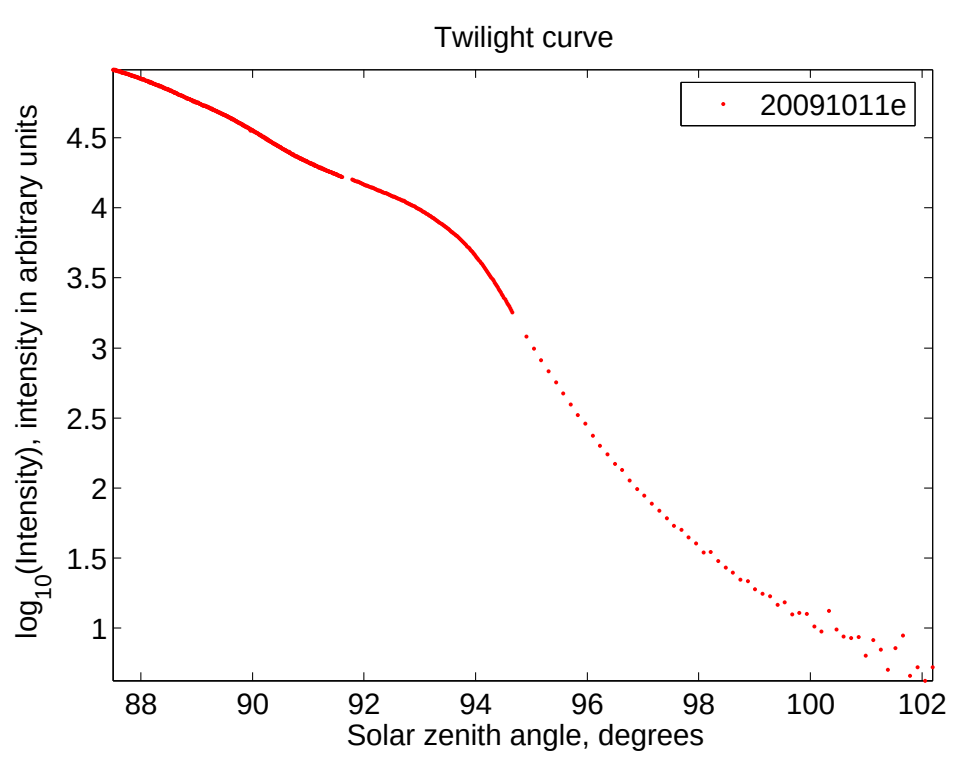
In this paper we present stratospheric and upper tropospheric aerosol extinction profiles retrieved from ground-based spectral measurements of twilight sky brightness.

It is well-known that stratospheric aerosol manifests itself by so-called "**purple light**", reddening of the twilight sky when the Sun is a few degrees below the horizon (**Mateshvili et al., 2005, JGR**). This effect is especially strong after major volcanic eruptions when the aerosol loading in the stratosphere increases dramatically. Photometrical measurements of the twilight sky brightnesses at one or more wavelengths are able to give some quantitative estimates of stratospheric aerosol loading.

2.Equipment

The measurements were carried out in the period from August 2009 to January 2011, in Tbilisi, Georgia (41° 43' N, 44° 47' E). We used a SBIG ST-9XE CCD camera equipped with SBIG SGS grating spectrograph. The field of view was 4°. The spectrograph was adjusted to take spectra between 700 and 800 nm. The spectral resolution was 3.8 nm.

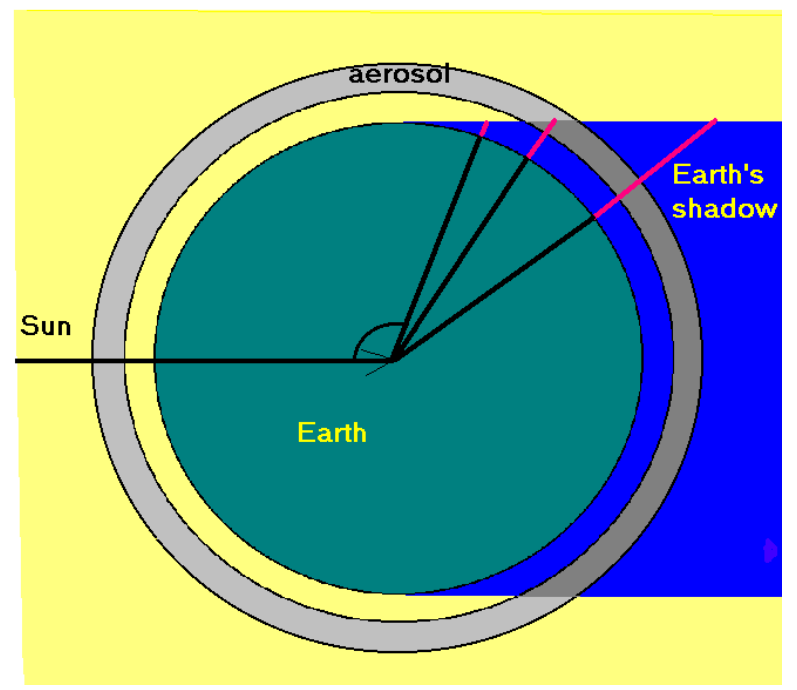




3. An example of the measured twilight sky light intensity.

The twilight curve is composed from three pieces which differ in the exposure time. The measured spectrum was averaged in the range 777-783 nm. The gaps between the ensembles correspond to the interruptions needed to change the exposure time. **The « hump » or « first purple light » between 92° and 94° SZA (solar zenith angle) is a stratospheric aerosol signature**.

4. Twilight event as a natural possibility of atmospheric sounding.

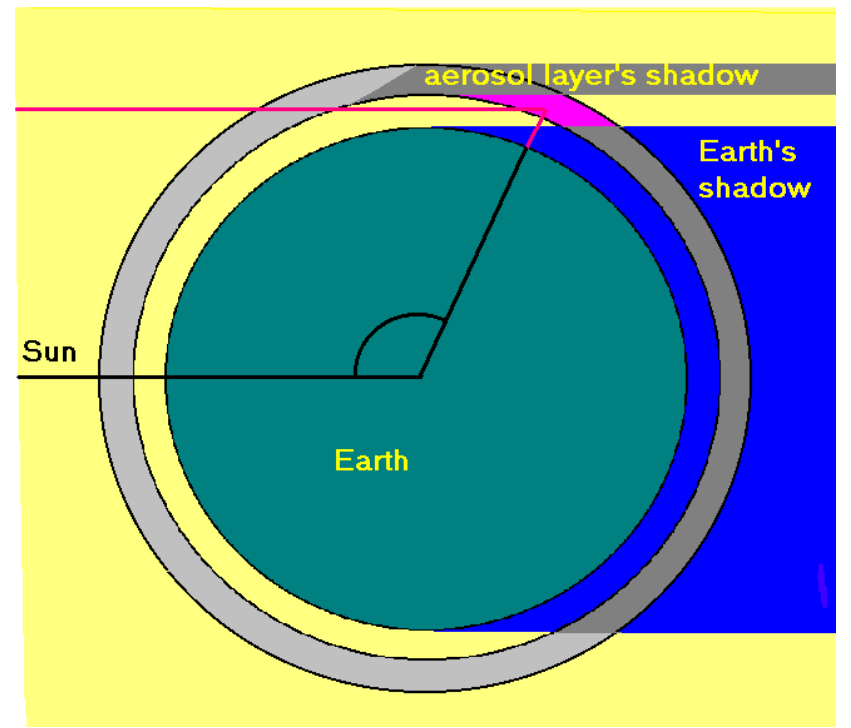


During twilight when the SZA is greater than 90° the atmosphere above the observer is partly shadowed and partly sunlit. The boundary between shadowed and sunlit parts of the atmosphere shifts with the progress of twilight leaving the lower layers of the atmosphere in the Earth's shadow. This gives us a natural possibility of atmospheric sounding.

When the Sun is below the horizon only the scattered light can be registered by the spectrometer. The intensity of the scattered light depends on vertical extinction profiles of different atmospheric species, as well as aerosol and molecular phase functions.

5. The stratospheric aerosol signature – first purple light

First purple light is a reddening of the sky western part (in the case of evening twilight) when the sun is a few degrees under the horison. Solar rays traveling a long way through the stratospheric aerosol layer at SZA greater then 90° are strongly attenuated and do not contribute significantly to the scattered light. Only those rays which cross the stratospheric aerosol layer at lower tangent heights and pass mostly under the layer may give significant contribution to the scattered light. This scattered light is observed in certain range of SZA and creates the hump on the twilight curve.



6. The forward model

The best choise of the forward model for the twilight problem is a 3-D Monte Carlo code. But such a code is too time-costing to be used directly as a forward model. We use the **Monte Carlo code Siro (Oikarinen et al.,1999)** to construct a simplified forward model where only single scattering is calculated and multiple scattering is considered as proportional to the single scattering. The coefficient of proportionality $\mathbf{F_{multiple}}$ is calculated using the Monte Carlo code

$$I_{total} = I_{single} F_{multiple} \quad (1)$$

where $\mathbf{I_{total}}$ is a total measured light intensity, $\mathbf{I_{single}}$ is a single scattering light intensity and $\mathbf{F_{multiple}}$ is a multiple scattering correction.
Fig. **a) (below)** shows $F_{multiple}$ variations with solar zenith angle for two aerosol extinction profiles presented on Fig. **b**. $F_{multiple}$ varies significantly with the aerosol profile variations that does not allow to use the expression (1) directly as a forward model.
The following transformation of measurements and the forward model was introduced:

$$\frac{d}{d(sza)}\log\left(I_{total}\right) = \frac{d}{d(sza)}\log\left(I_{single}\right) + \frac{d}{d(sza)}\log\left(F_{multiple}\right) \quad (2)$$

where SZA is solar zenith angle.

Fig. **c)** shows that after the transformation introduced in (2) the single scattering contribution starts to play a major role and the multiple scattering contribution can be considered as a weakly dependent from the actual aerosol profile. The reason why the transformation makes the multiple scattering correction relatively unsensitive to aerosol profile variations is that the multiple scattering correction changes slowly with solar zenith angle till SZA=94°. It should be mentioned that as the logarithmic derivative introduced in (2) removes all constant factors the absolute calibration is unnecessary.

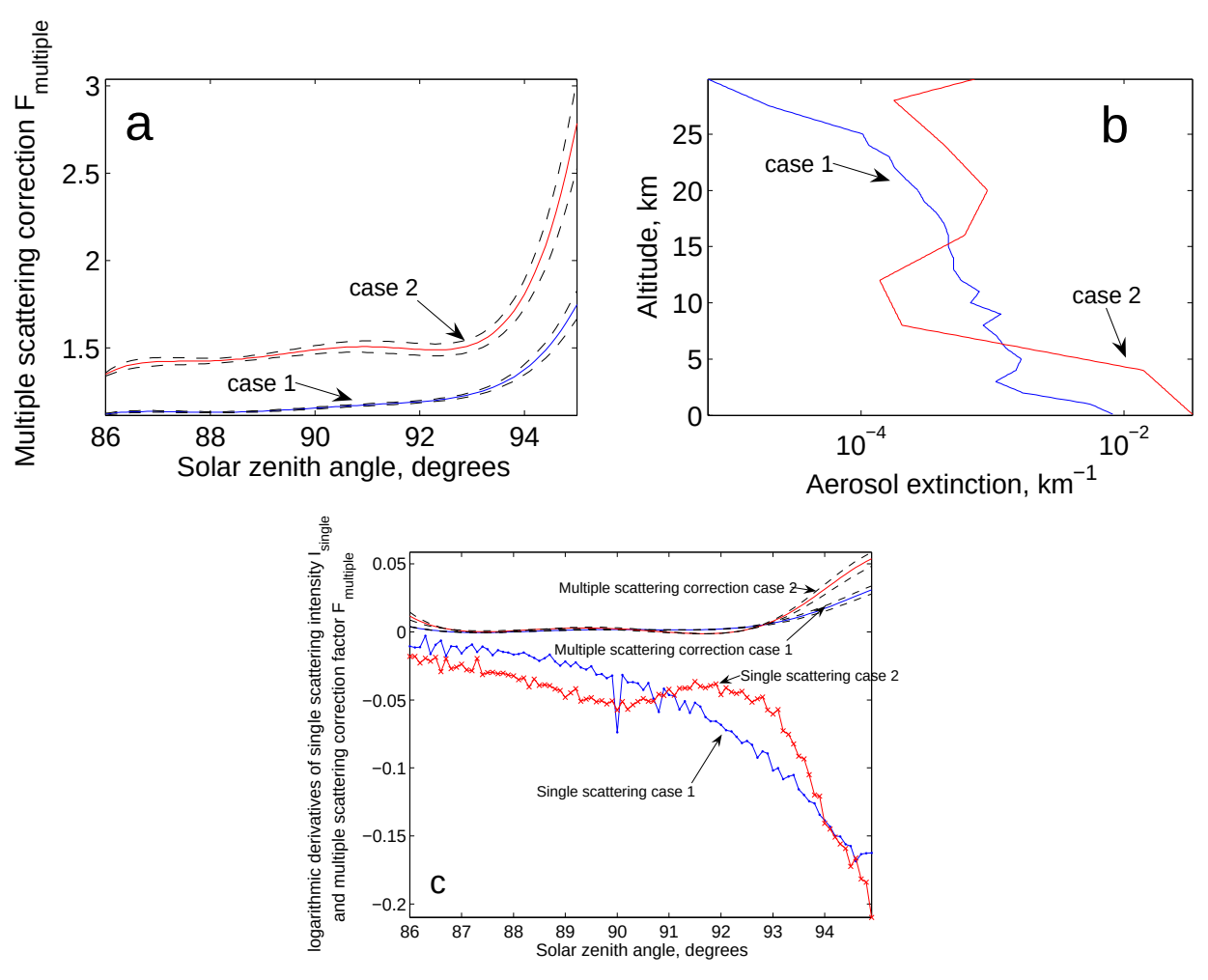
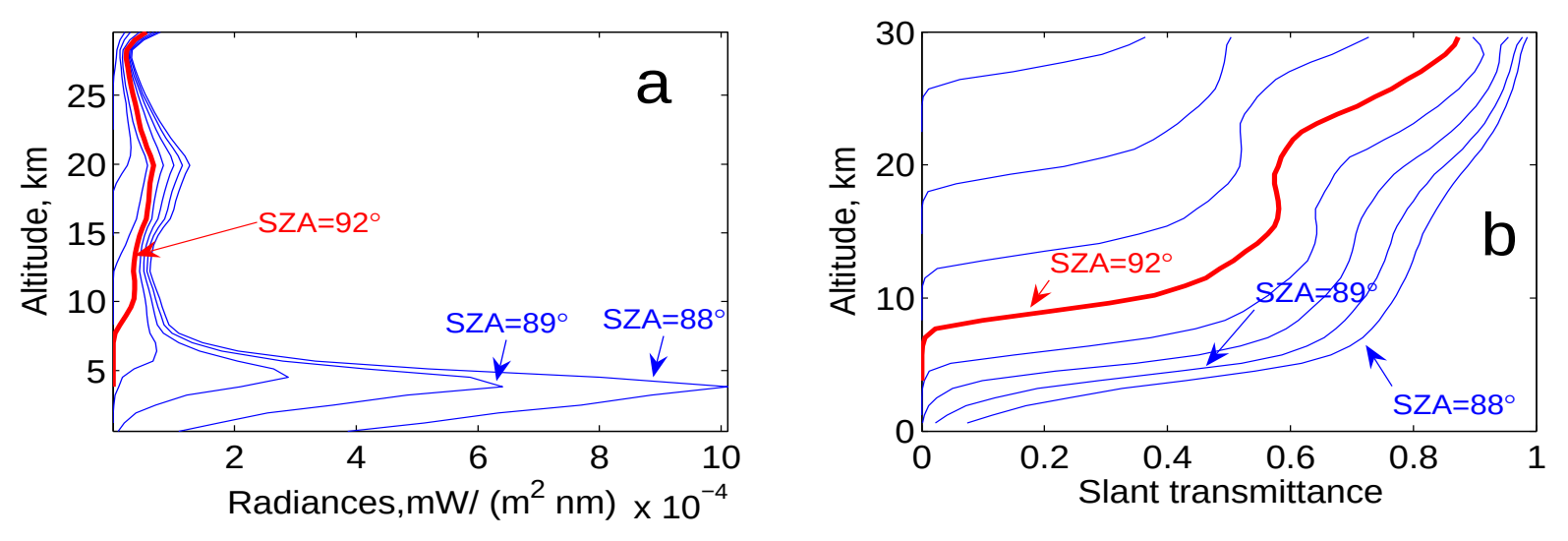


Figure left. a) Solar zenith angle (SZA) dependences of multiple scattering correction factors for two cases of aerosol extinction profiles presented on **(b**). Dashed line represent Monte Carlo uncertainties. **c)** Logarithmic derivatives for single scattering contribution and multiple scattering contribution modeled with the aid of Monte Carlo code for two cases of the aerosol extinction profiles presented on Fig. b.

7. The modeled contribution to the total twilight light intensity of singly scattered light from different altitudes at different solar zenith angles (SZA).

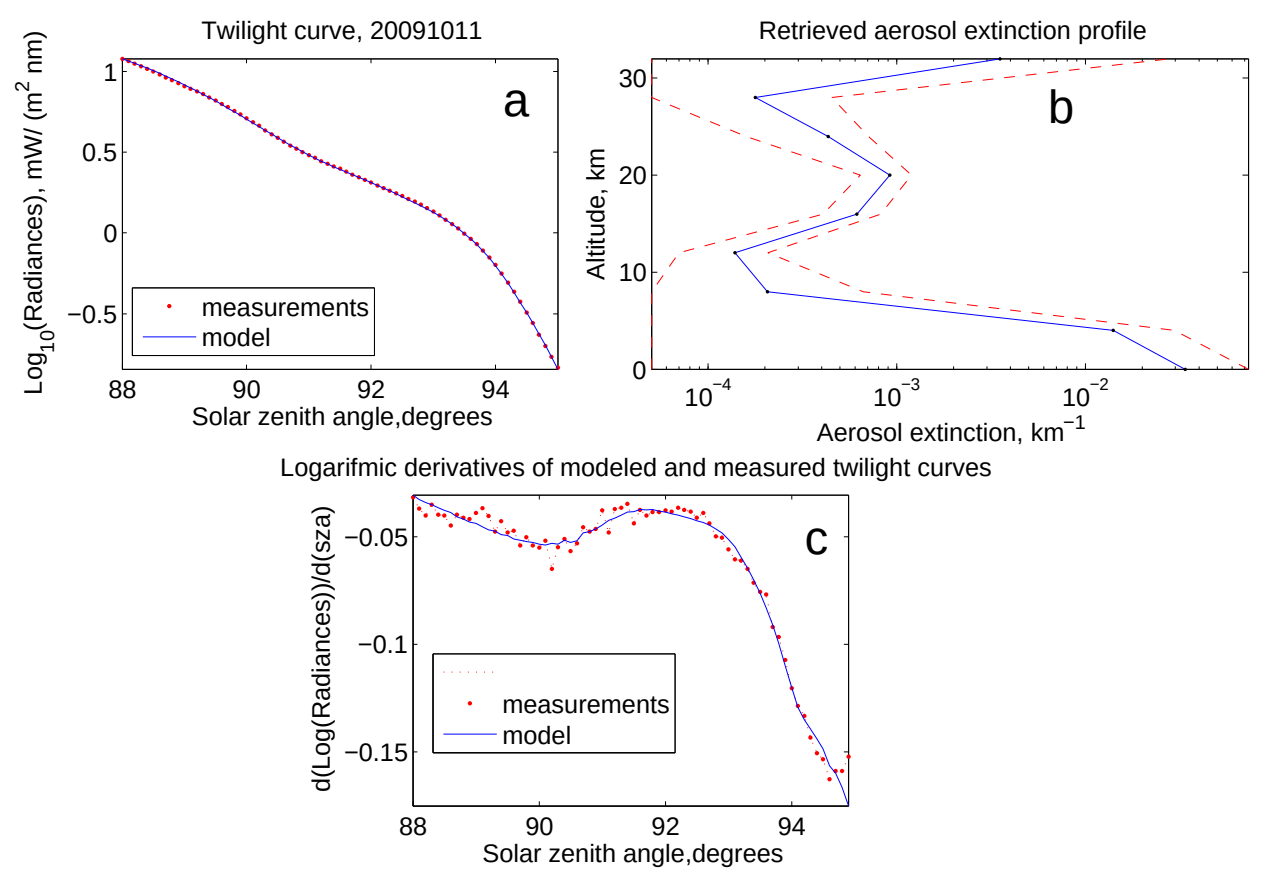
It is interesting to see how light scattered at different altitudes contributes to the total twilight light intensity at different solar zenith angles (SZAs). Fig. **a (below)** shows the altitudinal dependence of singly scattered light. It is clearly visible that at SZA 92° the tropospheric aerosol already does not contribute to the light intensity. Fig. **b** shows the corresponding slant path transmittances. The slant path's length increases with the increase of SZA and produces a transmittance cut-off in the lower part of the atmosphere. This is an important issue because **the retrieval results are independent from weather conditions at large distances from the observer.**



8. The retrieval example

The Levenberg-Marquardt algorithm was used to retrieve the aerosol extinction profiles. The US Standard Atmosphere was used to model the Rayleigh scattering. There is no absorption by atmospheric gases at the chosen wavelength (780 nm).

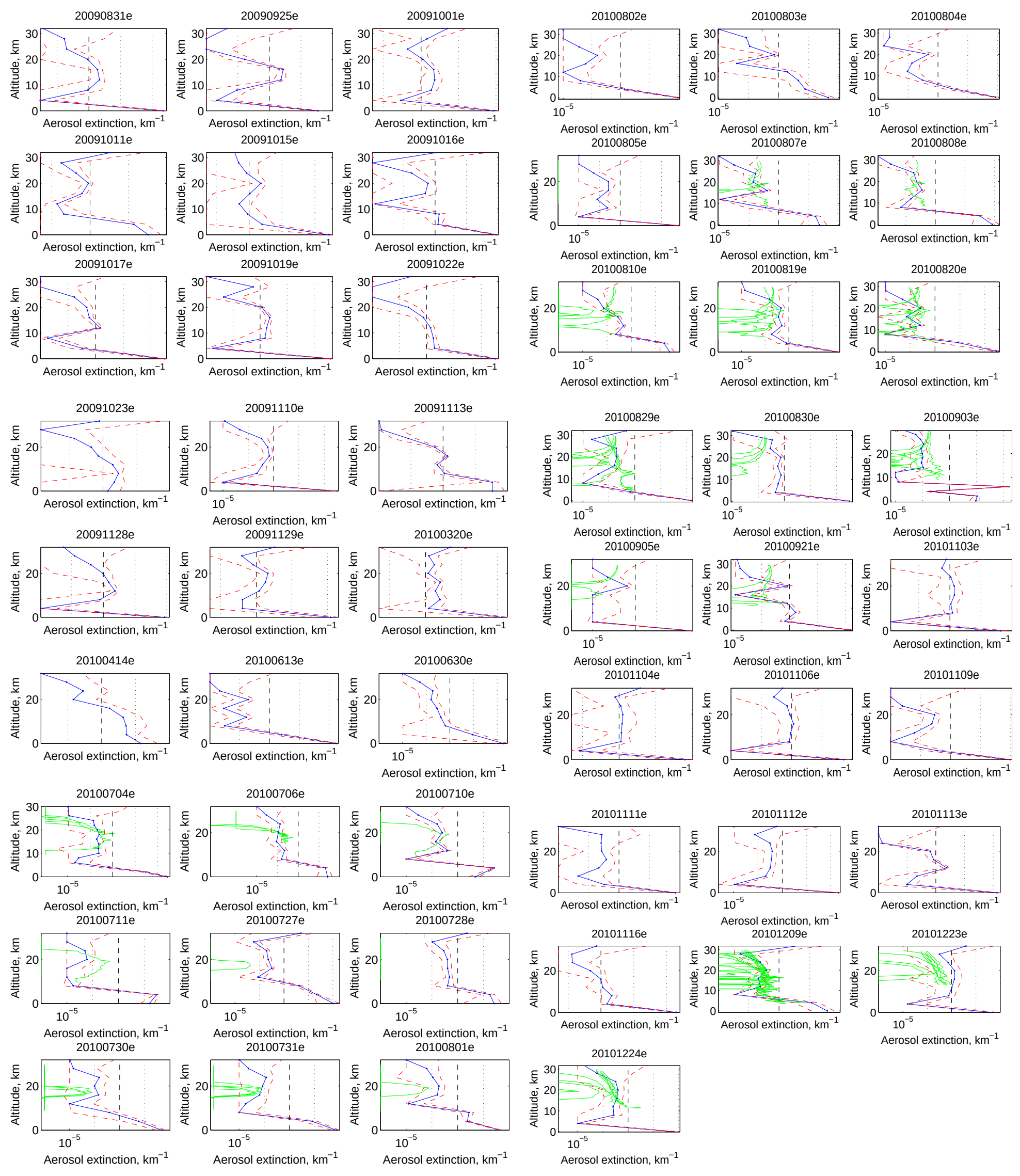
Figure **(right)** shows **a)** an example of the twilight curve fitted with the help of the forward model described in section 6. Blue curve is the model, red dots are data points , **b)** The aerosol extinction profile (blue) with it's uncertainties (red) retrieved from the twilight curve (a). **c)** The logarithmic derivative of the modeled (blue) and measured(red dots) twilight curves presented on (a).



9. The aerosol extinction profiles retrieved from twilight measurements and validated by GOMOS aerosol extinction profiles

The retrieved aerosol extinction profiles (blue) with the retrieval uncertainties (red dashed) and GOMOS (Global Ozone Monitoring by Occultation of Stars instrument onboard the ENVISAT satellite) aerosol extinction profiles(green).

Grid lines correspond to powers of 10 of the extinction . Dashed black line corresponds to the extinction 10^{-3} km^{-1}.



10. Stratospheric and tropospheric aerosol optical depth variations

Stratospheric **(a)** and tropospheric **(b)** aerosol optical depth variations from September 2009 to January 2011 integrated over the retrieved aerosol extinction profiles presented above. The increase of stratospheric aerosol loading during fall of 2009 is caused by Sarychev Peak eruption in the middle of June 2009.