Rotational Raman scattering in limb viewing geometry: modeling with SCIATRAN

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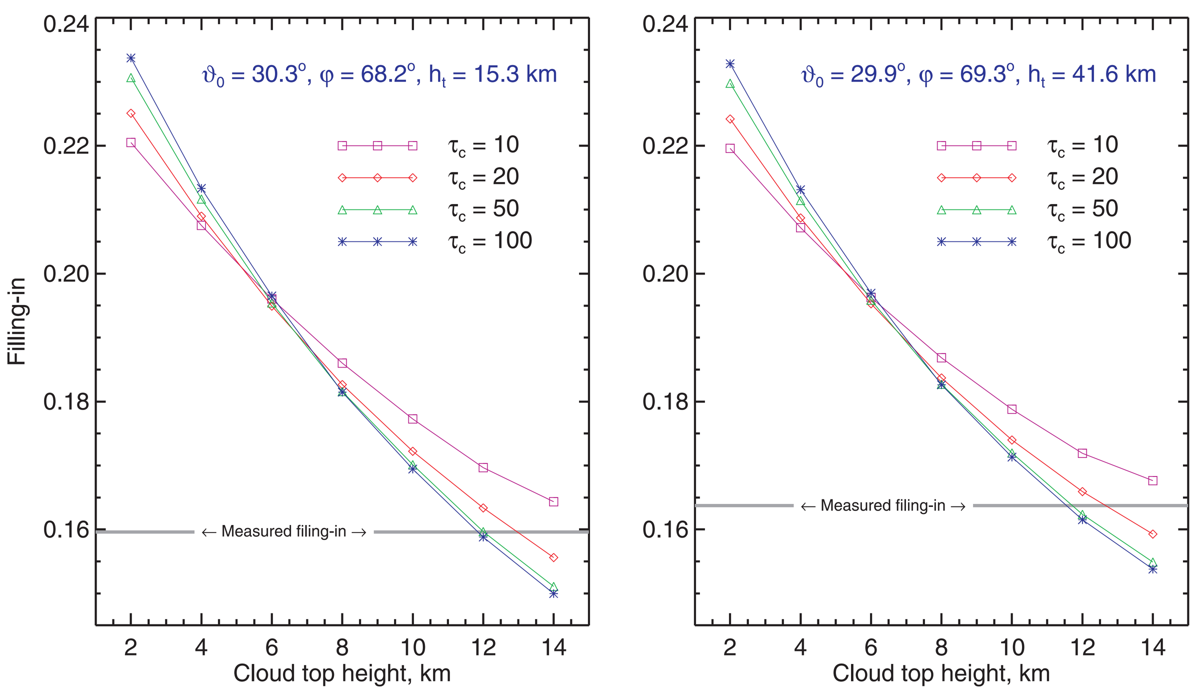
Introduction

The topic of this presentation is the modeling of the Ring effect in measurements of the scattered solar light performed in a limb viewing geometry. This effect has been discovered independently by *Shefov (1959)* and *Grainger and Ring (1962)* and is observed as a slight difference in the depths of the solar Fraunhofer lines in the spectra of the direct and the scattered solar light (so called filling-in). The magnitude of the filling-in is usually in the order of a few percent and is thus less important for the interpretation of the absolute radiance measurements. However, when differential absorption structure of atmospheric trace gases is used in the retrieval process (DOAS approach) the influence of the Ring effect might be crucial. In addition, the magnitude of the solar Fraunhofer lines filling-in can be used to gain information about the cloud properties in the underlying scene.

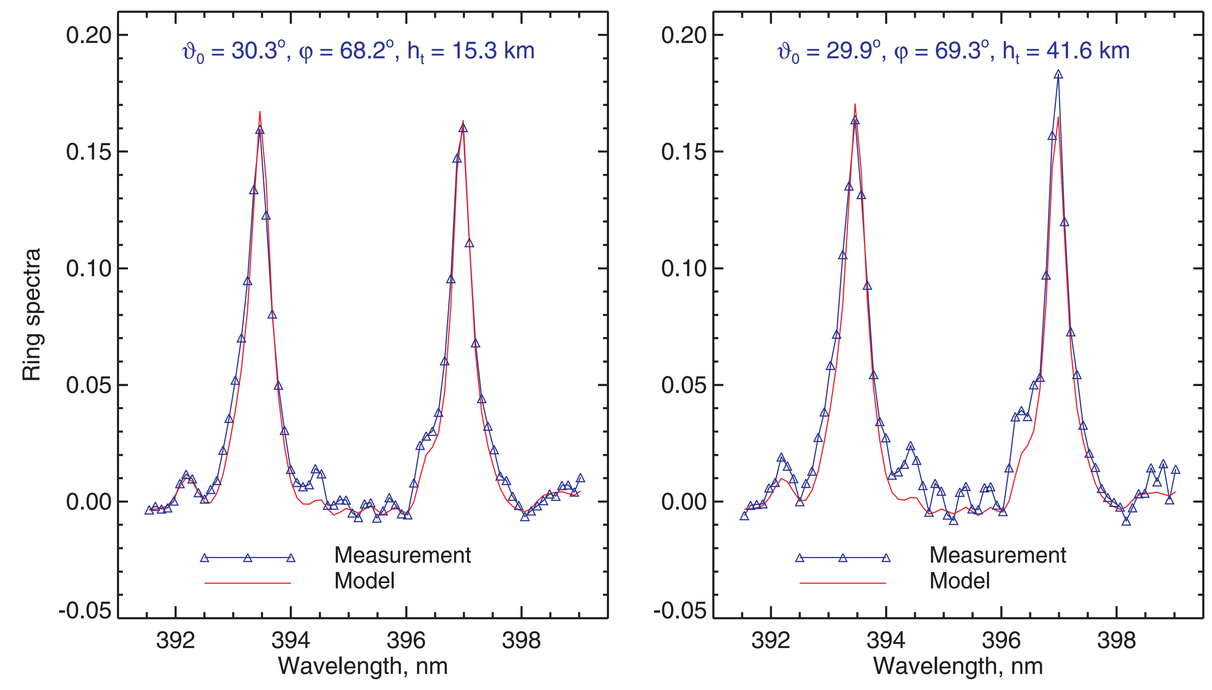
Currently, the scientific community concurs that the Ring effect is explained by the rotational Raman scattering by nitrogen and oxygen molecules occurring in the Earth's atmosphere. The Raman scattering is an inelastic scattering process associated with a change of a photon energy as a result of the scattering event. The rotational Raman scattering re-distributes the energy of the scattered photons over several nanometers in the wavelengths domain. The loss of intensity is proportional to the local intensity of the radiation field whereas the gain is proportional to the intensity at neighboring wavelengths which results in a filling-in of deep Fraunhofer lines and absorption signatures of trace gases in measured spectra of scattered solar radiation.

The radiative transfer model SCIATRAN 3.2 (*Rozanov et al., 2013*) has been recently extended to account for the rotational Raman scattering in limb viewing geometry. The Raman scattering by N_2 and O_2 is accounted for. The solution is obtained employing characteristic and discrete-ordinates methods in combination with forward-adjoint technique. Only the first order of scattering in the wavelength domain is accounted for. The results obtained with the model are presented here.

RRS spectra over cloudy scenes



Filing-in magnitude for Ca II Fraunhofer line at 393.461 nm as a function of the cloud top height modeled with SCIATRAN for a cloud geometrical thickness of 1 km and different optical thicknesses. Calculations were performed including convolution with Gaussian slit function (FWHM = 0.14 nm). The solid horizontal line without symbols denoted as "Measured filling-in" represents the filling-in as calculated from SCIAMACHY measurements. Results for tangent heights 15.3 km and 41.6 km are presented in the left and right panel, respectively.

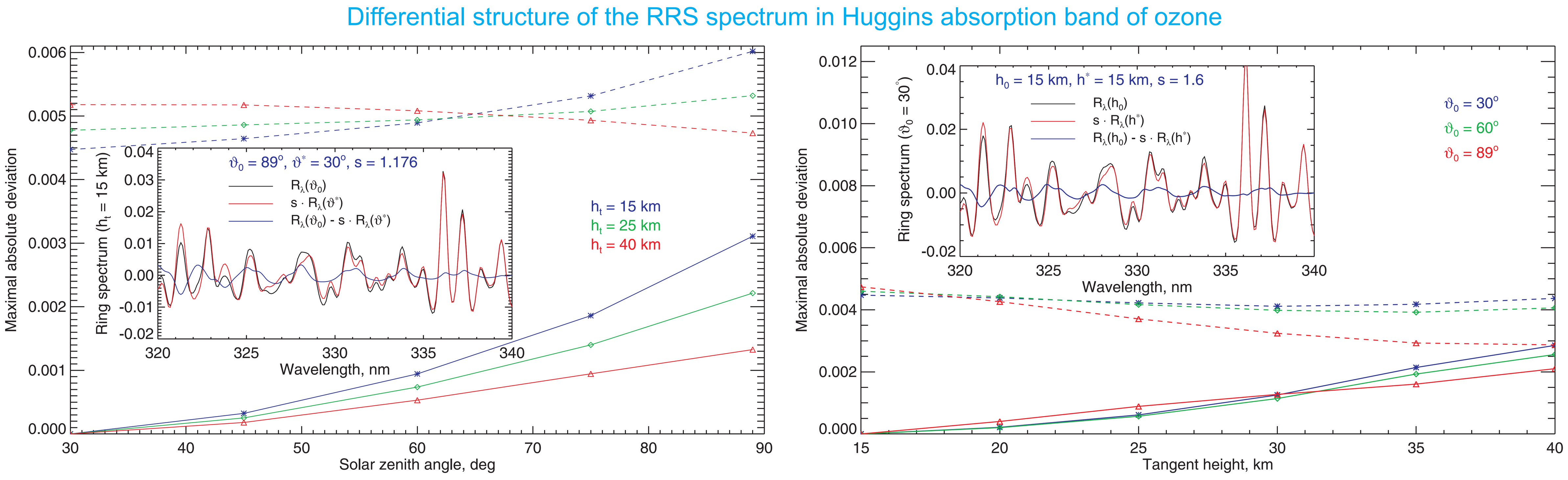


Ring spectra in the spectral range 391-399 nm at two tangent heights, 15.3 km (left) and 41.6 km (right), derived from SCIAMACHY observations in the limb-viewing geometry (blue) and modeled with SCIATRAN (red). The model calculations were done for a water cloud with an optical thickness of 50 between 11 and 12 km. The cloud was assumed to consist of spherical particles with an effective radius of 6 μm. The results remain the same if an ice cloud consisting of fractal particles of the second generation on the base of a regular tetrahedron with effective radius 23 μm is assumed instead.

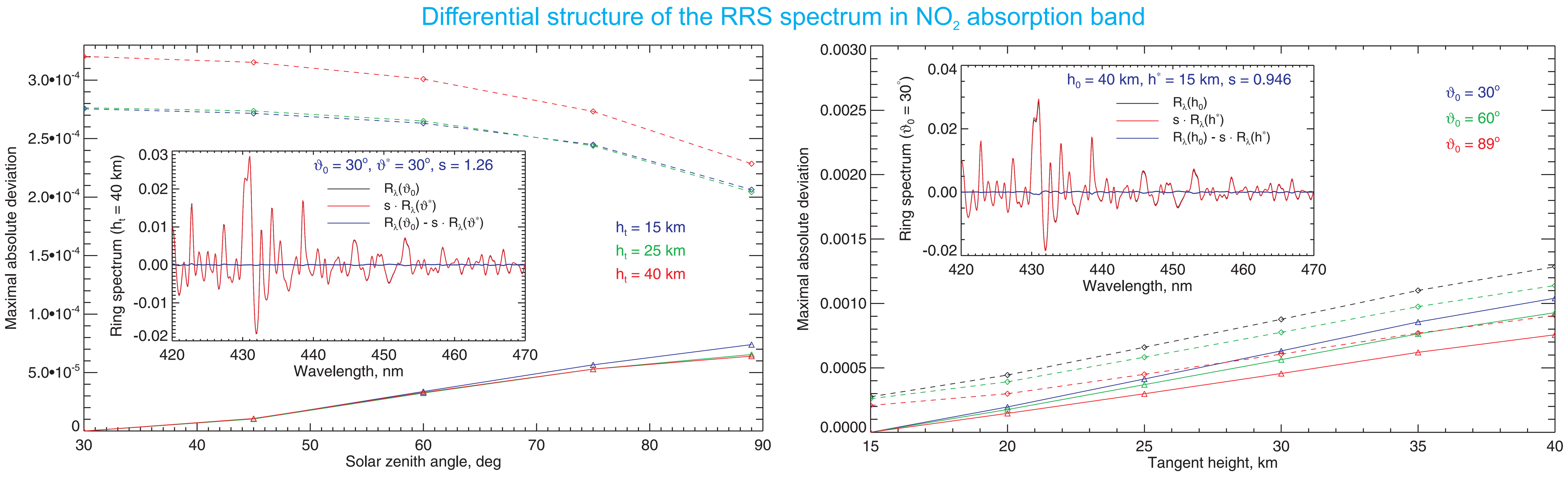
Using the rotational Raman scattering (RRS) spectrum for DOAS applications

Interpretation of limb-scatter observations is often done employing the so-called DOAS technique which exploits the differential absorption structure of atmospheric trace gases to gain information about their number densities. If the sun-normalized radiance is used for the retrieval the influence of the RRS can be quite high and must be accounted for. As a modeling of RRS spectra is quite time consuming, a spectrum pre-calculated for any selected observations conditions is commonly used in the fit procedure. The figures below illustrate the maximum errors arising from this kind of fit for different spectral ranges. In the left panels the RRS spectrum was calculated for a solar zenith angle of 30° and fitted then to other solar zenith angles. This was done for tangent heights of 15, 25 and 40 km. The solid lines show the results obtained accounting for the multiple scattering in all RRS spectra, while the dashed lines illustare the fit quality is the first RRS spectrum (which is fiited to all others) is calculated in the single scattering approximation. The small plots inside the main panels illustrate the modeled and fitted RRS spectra and their difference for the worst case geometry. The geometry of the original spectrum is marked with "*" and that of the fitted spectrum with "0" while "s" denotes the fit coefficient. The right panels show similar results but with RRS spectrum simulated for a tangent height of 15 km and fitted then to other tangent heights. The comparison is done for solar zenith angles of 30°, 60° and 89°. All calculations are done for a relative azimuth angle of 30°.

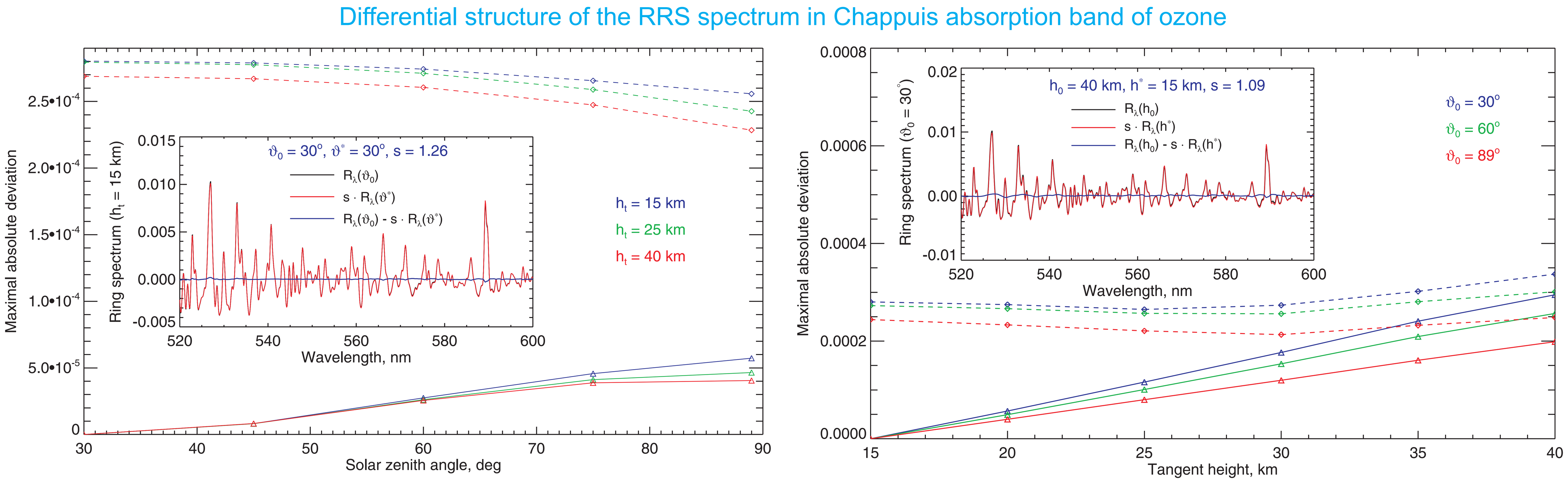
Differential structure of the RRS spectrum in Huggins absorption band of ozone



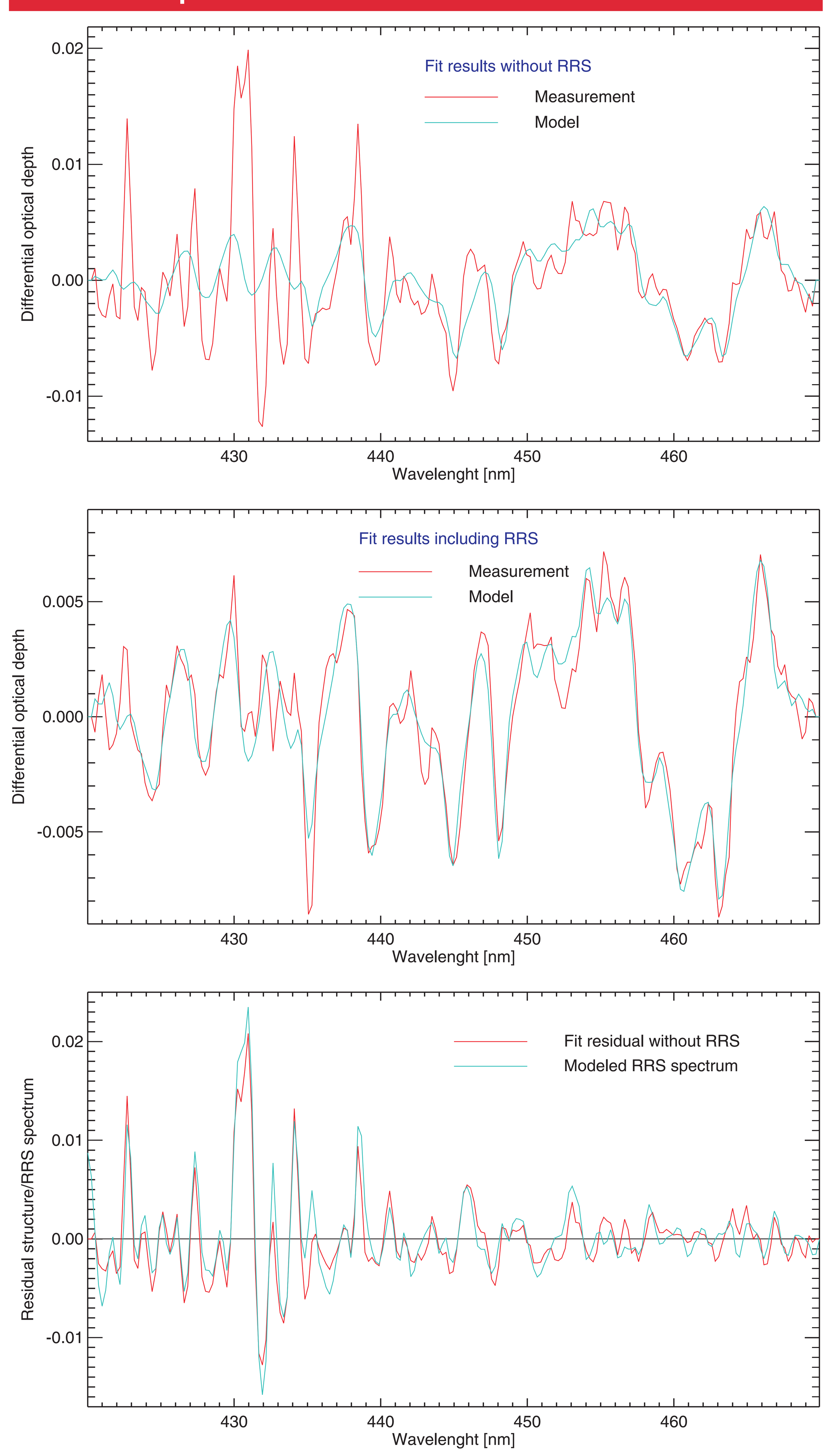
Differential structure of the RRS spectrum in NO_2 absorption band



Differential structure of the RRS spectrum in Chappuis absorption band of ozone



Copmarison to measurements



Upper and middle panes: spectral fits neglecting the RRS contribution and including the RRS spectrum as a pseudo-absorber in the DOAS fit procedure. The fit includes spectral signatures of O3, NO2, O4, instrumental corrections and a cubic polynomial. Red curve shows the measured spectrum and cyan curve represents the model simulation. Lower panel: residual form the fit shown in the upper panel (red) in comparison with a RRS spectrum modeled by SCIATRAN (cyan). The comparison is done for a SCIAMACHY limb measurement at a tangent height of 12 km. The measurement was performed at a solar zenith angle of 45° and a relative azimuth angle of 50° (angles are given at a tangent point).

References

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