

# Correction of the Ring effect and $I_0$ -effect for DOAS observations of scattered sunlight

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## Introduction

Absorption spectroscopy of scattered Sun light has become a widely used method for the measurement of atmospheric trace gases during the last century [Dobson and Harrison, 1926; Brewer et al., 1973; Noxon et al., 1975; Solomon et al., 1987; Burrows et al., 1988; Platt et al., 1997].

Such observations are affected by two principal effects:

A) The contribution of inelastically scattered light (Raman scattering) to the total intensity leads to a modification which can be approximated as filling in of the Fraunhofer lines. This effect complicates the complete removal of the Fraunhofer structures: it is usually called Ring effect [Grainger and Ring, 1962].

B) The limited spectral resolution of a typical DOAS instrument leads to an interference of the Fraunhofer structures of the solar spectrum and the absorption cross sections of the atmospheric trace gases [Johnston, 1996].

For the correction of both effects substantial progress has been made during the last years. An overview on the discovery, the history of the correction methods and the current status is given in this presentation.

## Observations of scattered sun light

Absorption spectroscopy of atmospheric trace gases is based on the Beer-Lambert law:

$$I(\lambda) = I_0(\lambda) \cdot e^{-\sigma(\lambda)\rho l} \quad (1)$$

$I(\lambda)$  indicates the measured spectrum,  $I_0(\lambda)$  the unattenuated intensity,  $\sigma$  the absorption cross section,  $\rho$  the trace gas concentration and  $l$  the length of the absorption path.  $\sigma \cdot \rho \cdot l = \tau$  (optical depth).

To remove the strong Fraunhofer structures in the measured spectrum  $I(\lambda)$  can be divided by another measured spectrum (direct sun light or atmospheric spectrum at low SZA; this spectrum is often called Fraunhofer spectrum), which will be referred to as  $I_0(\lambda)$  in the following. The optical depth  $\tau$  of the atmospheric absorption is then derived by taking the negative logarithm of the ratio spectrum.

$$\sigma(\lambda) \cdot l \cdot \rho = \tau(\lambda) = -\ln \frac{I(\lambda)}{I_0(\lambda)} = -\ln I(\lambda) + \ln I_0(\lambda) \quad (2)$$

For DOAS applications the spectra and cross sections are usually high pass filtered and the differential quantities  $\sigma'$  and  $\tau'$  are used. Often the logarithm of the Fraunhofer spectrum is fitted (together with the cross sections of the trace gases) to the logarithm of the measured spectrum.

$$\left| \sum_i \ln I(\lambda_i) - \sum_i \ln I_0(\lambda_i) - \sum_k \alpha_k \cdot \sigma'_k(\lambda_i) - \sum_j \lambda_i^j \right| \rightarrow 0 \quad (3)$$

$\sigma'_k(\lambda_i)$  denotes the differential cross section of the  $k$ -th trace gas taken into account in the fitting process. The derived fit coefficient  $\alpha_k$  yields the desired SCD of this species.

$\sum_j \lambda_i^j$  is a polynomial often used to account for broad band features, e.g. from Rayleigh- and Mie-scattering.

### A) The Ring effect

The measured spectrum (and in many cases also the Fraunhofer spectrum) is the sum of elastic scattering processes ('Rayleigh-' and 'Mie-scattering') and inelastic scattering processes (Raman scattering):  $I = I_{\text{elastic}} + I_{\text{inelastic}}$  [see e.g. Young, 1981]. Thus by taking the ratio of these spectra (eq. 2) the Fraunhofer structures of the measured spectrum are not fully removed. Especially in the UV spectral range the remaining spectral structures (up to 10%) can thus by far exceed the absorption structures of weak atmospheric absorbers (like e.g. BrO and OCIO). While at the beginning of atmospheric UV/vis absorption measurements the Ring effect was not corrected for, today sophisticated methods for a proper correction are usually applied.

## Historical overview

- Grainger and Ring [1962], Discovery of the Ring effect.
- Brinkmann [1968], Kattawar et al. [1981], **Rotational Raman** scattering most likely cause for Ring effect.
- Noxon et al. [1979], Ring effect causes negligible residual structures; they can be reduced by adding a constant offset (<2%) to the measured spectrum before ratioing.
- McKenzie and Johnston [1982], Residual structures are mainly caused by imperfect removal of Fraunhofer structures
- Solomon et al. [1987], **Ring spectrum** from polarisation measurements **included in fit process**.  
This procedure proved to be very successful and was adopted by many groups [Wahner et al., 1990; Perner et al., 1991; Fish and Jones, 1995; Richter, 1997]
- Pommereau and Goutail [1988], Spectral ranges of strong Fraunhofer lines are simply ignored.
- Johnston and McKenzie [1989], Reciprocal of the Fraunhofer spectrum used as Ring spectrum.
- Bussemer [1993], **Calculated Ring spectrum** using single scattering atmospheric radiative transport model RASCAMF.  
Since then several similar or improved codes were used by several groups [Fish and Jones, 1995; Vountas et al., 1998; Sioris and Evans, 2000; Aben et al., 2001].
- Van Roozendaal et al. [1995], Ring spectrum from high pass filtering of the Fraunhofer spectrum.
- Fish and Jones [1995], Errors of retrieved trace gas amounts due to **Filling in of atmospheric** absorption lines (NO<sub>2</sub>). Similar investigations for the O<sub>2</sub> A band were performed by Sioris and Evans [2000].
- Joiner and Bhartia [1995], Determination of cloud top height from the magnitude of the Ring effect (see also DeBeek et al. [2001]).
- Haug [1996], Investigation of the possible impact of **vibrational Raman scattering** on atmospheric measurements.
- Vountas et al. [1998], Principle component analysis of Ring spectra, **separation of Filling in of Fraunhofer lines and atmospheric absorption lines**.
- Chance [1998], ‘Direct intensity fitting’: Replacement of Ring spectrum by Raman intensity spectrum (Results are comparable to those of ‘classical DOAS’)

- Wagner [1999], Correction of the broad band spectral dependence of the Ring effect (for different atmospheric scenarios) using two Ring spectra.
- Aben et al. [2001], Measurement and modelling of the polarisation due to the Ring effect.

### Correction of the Ring effect

Usually the Ring effect can be approximately corrected for by considering an additional spectrum (ore more than one additional spectrum, see below) in the DOAS fitting process, the so called Ring spectrum. This approach was introduced by Solomon et al. [1987]; only after this important step in the correction of the Ring effect it was possible to observe ‘minor atmospheric absorbers’ (like BrO and OCIO) with absorptions  $\ll 1\%$ .

The logarithm of the measured spectrum (eq. 3) can be approximated:

$$-\ln(I_{meas}) = -\ln(I_{elastic} + I_{inelastic}) \approx -\ln(I_{elastic}) + \frac{I_{inelastic}}{I_{elastic}} \quad (4)$$

with

$$I_{Ring} := I_{inelastic}/I_{elastic} \quad (5)$$

$I_{Ring}$  is included in the spectral fitting process (eq. 3). It can be either calculated or measured [Solomon et al., 1987; Bussemer, 1993].

It should be noted here that besides the ‘classical’ DOAS (fitting the optical depth) also methods are developed which directly fit the measured intensities [Chance 1998]. In such cases the Raman scattered intensity (instead of the ‘classical’ Ring spectrum [Solomon et al., 1987]) is included as a Ring spectrum in the fitting process.

Another method to account for the influence of Raman scattering upon the measured spectra is proposed by Kostadinov et al. [1997]. This method performs a deconvolution of the measured spectrum using a previously built ‘Atmospheric Slit Function’ (ASF), containing both Rayleigh and Raman components. The construction of ASF allow to take into account the atmospheric temperature and to include the atmospheric constituents, which contribute to the scattering process.

### Comparison of Ring spectra of different groups for the spectral range 340 to 370 nm

In Figure 1 Ring spectra used in various groups are shown for the wavelength range 340 - 370 nm. Although the spectra are calculated for different spectral resolution using different techniques the spectral structures are very similar in the different Spectra.

In order to allow a more meaningful comparison between the different methods selected Ring spectra from Fig. 1 are shown in Figs. 2 and 3. (in Fig. 2 some of them have been convolved to achieve comparable spectral resolution). While similar structures are present in all three spectra and in some cases the agreement is nearly perfect, significant differences also appear. In particular for the comparison in Fig. 2 the structures at 344 and 359 nm have different magnitudes: For the Ring spectrum derived from polarisation measurements the structure at 344 nm is about 51% of that at 359 nm; for the calculated one it is about 88%. In Fig. 3 the structures are very similar (for the calculated one about 82% and for the measured one about 84%).

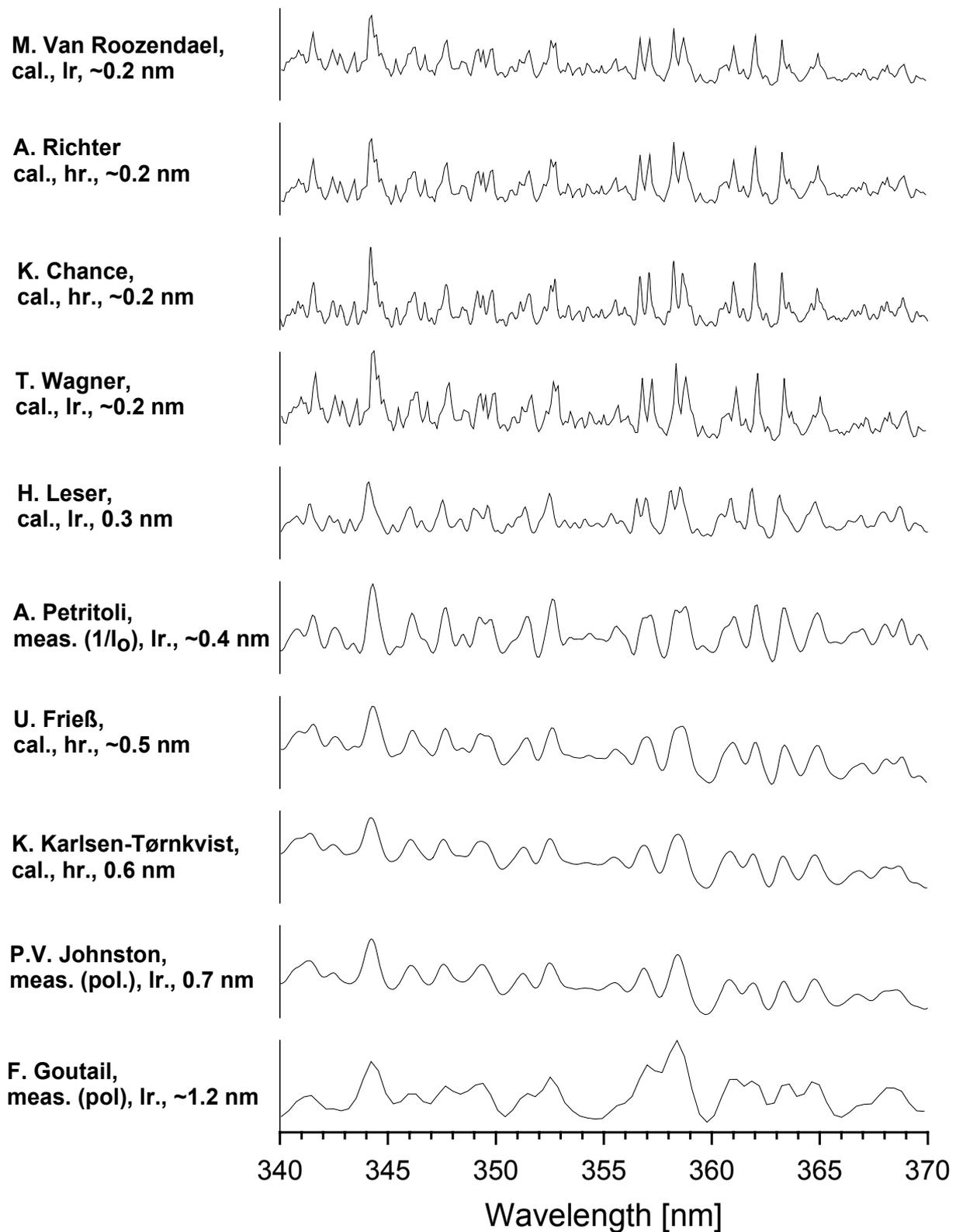


Fig. 1 Comparison of different Ring spectra used in different groups.

- meas./cal.: The Ring spectrum was measured or calculated;
- pol.: Polarisation measurements were used to determine the Ring spectrum
- 1/I<sub>0</sub>: The Reciprocal of a measured spectrum is used
- hr./lr.: A sun spectrum with high or low resolution was used.
- x nm: Approximate spectral resolution of the spectrum (FWHM)

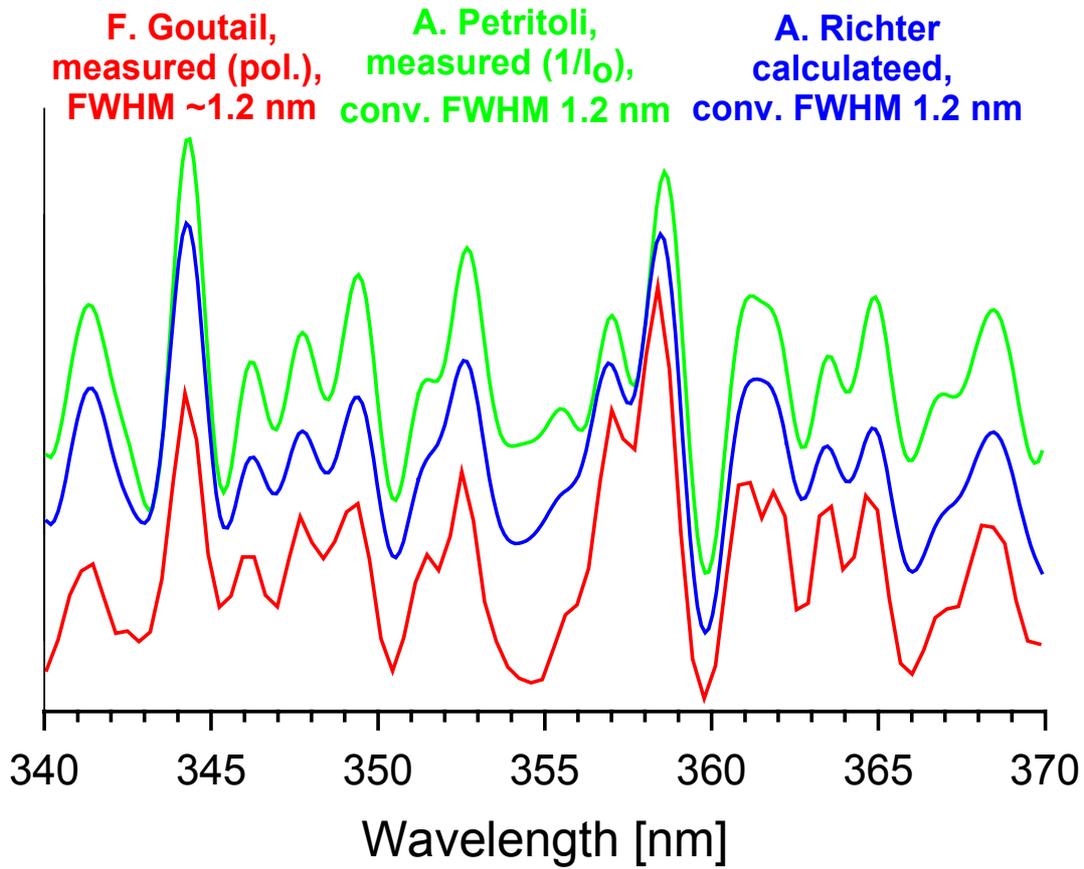


Fig. 2 Comparison of selected Ring spectra from Fig. 1 (for a better comparison the Ring spectra from A. Petritoli and A. Richer have been convolved with a Gaussian function of a FWHM of 1.2 nm).

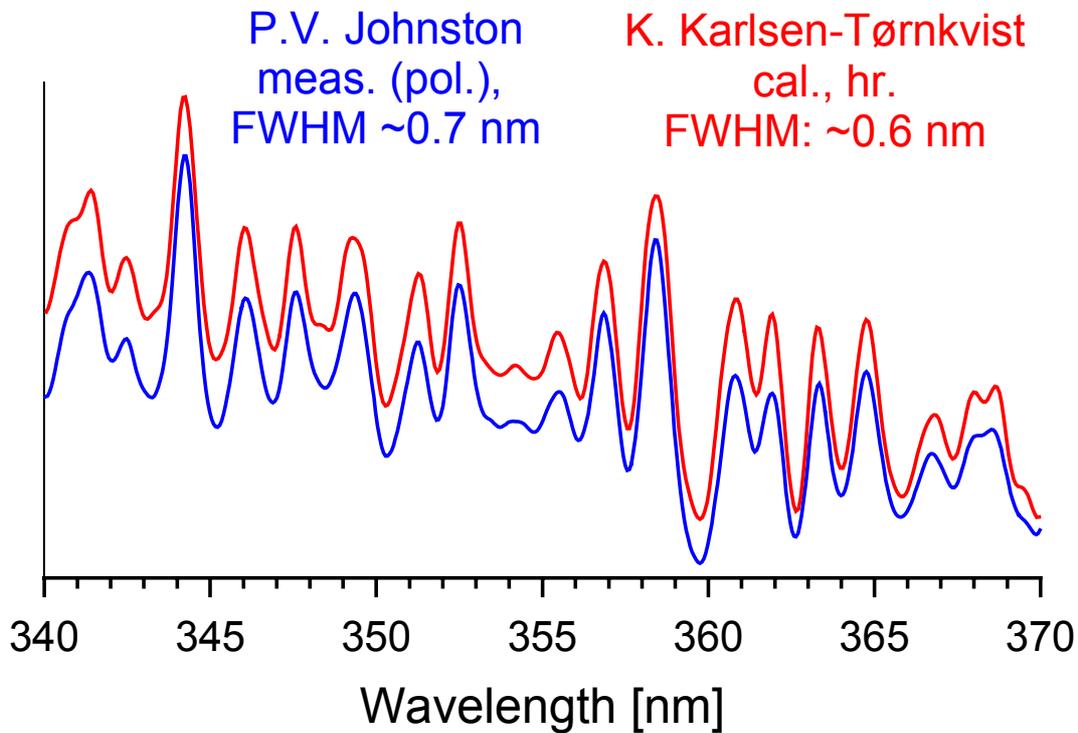


Fig. 3 Comparison of selected Ring spectra from Fig. 1 with similar spectral resolution.

## **Current status of the Ring correction in several groups**

### **Determination of Ring spectra**

Currently most of the groups calculate Ring spectra (assuming Raman scattering as cause of the Ring effect). One group uses the reciprocal of the measured spectrum (assuming that the Raman scattered intensity is constant with wavelength); three groups use measured Ring spectra (either from polarisation measurements or from a combination of direct and scattered sun light observations). Please note that not for all of these methods Ring spectra could be provided (Figs. 1, 2 & 3).

One problem of a measured Ring spectrum is that it usually contains absorption structures of atmospheric trace gases, because of the different light paths for cross polarised light (or direct and scattered sun light). These remaining structures of atmospheric absorbers in measured Ring spectra affect the analysis of the atmospheric measurements.

The higher aerosol load in the northern hemisphere might be one reason why northern hemisphere researchers prefer calculated Ring spectra.

### **Sun spectrum used for the determination of the Ring spectra**

Some groups use highly resolved solar spectra [Kurucz et al., 1984; Hall and Anderson, 1991], some groups prefer spectra measured by their own instrument (either containing atmospheric absorptions or direct sun spectra from satellite). The use of high resolved sun spectra should yield the more accurate Ring spectra (eq. 4,5). However, sometimes the convolution with a not perfectly known instrument function limits the quality of the Ring spectrum.

### **Atmospheric parameters for the determination of the Ring spectrum**

Most groups don't account for changes of solar zenith angle (SZA), atmospheric temperature and pressure. One group derives an individual Ring spectrum for each observation. While it is possible to model the full radiative transport [see e.g. Bussemer, 1993; Vountas et al., 1998; Aben et al., 2001] most groups calculate Raman scattering only for average atmospheric conditions [Chance and Spurr, 1997].

### **Number of Ring spectra used in the fitting process**

There are two general reasons for considering more than one Ring spectrum in the fitting process:

a) Fish and Jones [1995] investigated the influence of Raman scattering on the filling in of atmospheric absorptions. They found out that in particular the absorptions of narrow absorption bands (e.g. of NO<sub>2</sub>) can be significantly underestimated. Vountas et al. [1998] correct for the filling in of atmospheric absorptions by calculating additional Ring spectra including the atmospheric absorptions. These spectra (often they are orthogonalised) are then included in the spectral fitting process (see Figure 4). Computer programs for the determination of sets of Ring spectra including atmospheric absorbers are available at the Belgian Institute for Space Aeronomy (Michel Van Roozendael, e-mail: Michel.Vanroozendael@oma.be) and at the Smithsonian Astrophysical Observatory (K. Chance, e-mail: kchance@cfa.harvard.edu). Ring Eigenvector spectra can also be received from Marco Vountas, e-mail: Marco.Vountas@iup.physik.uni-bremen.de.

b) The fraction of elastically scattered light (see eq. 4) shows very different broad band spectral dependencies:  $\approx\lambda^{-4}$  for Rayleigh scattering;  $\approx\lambda^{-1}$  for Mie scattering;  $\approx\lambda^0$  for reflection at the ground. Thus the relative contribution of Raman scattering to the filling in of Fraunhofer lines depends strongly on the atmospheric properties (clear/cloudy sky, high/low ground albedo, SZA). This dependence can be corrected for by using two Ring spectra with different broad band amplitudes (Figure 5) [Wagner, 1999].

### Problems, Suggestions

In general most groups are almost satisfied with their current status of the Ring correction. Some groups want to improve their Ring spectra by using more sophisticated models (e.g. more detailed atmospheric radiative transport calculations; use of high resolved solar spectra). Some groups report problems when performing off-axis measurements from the ground. In such cases the residuals caused by imperfect correction of the Ring effect are significantly larger than for zenith sky observations.

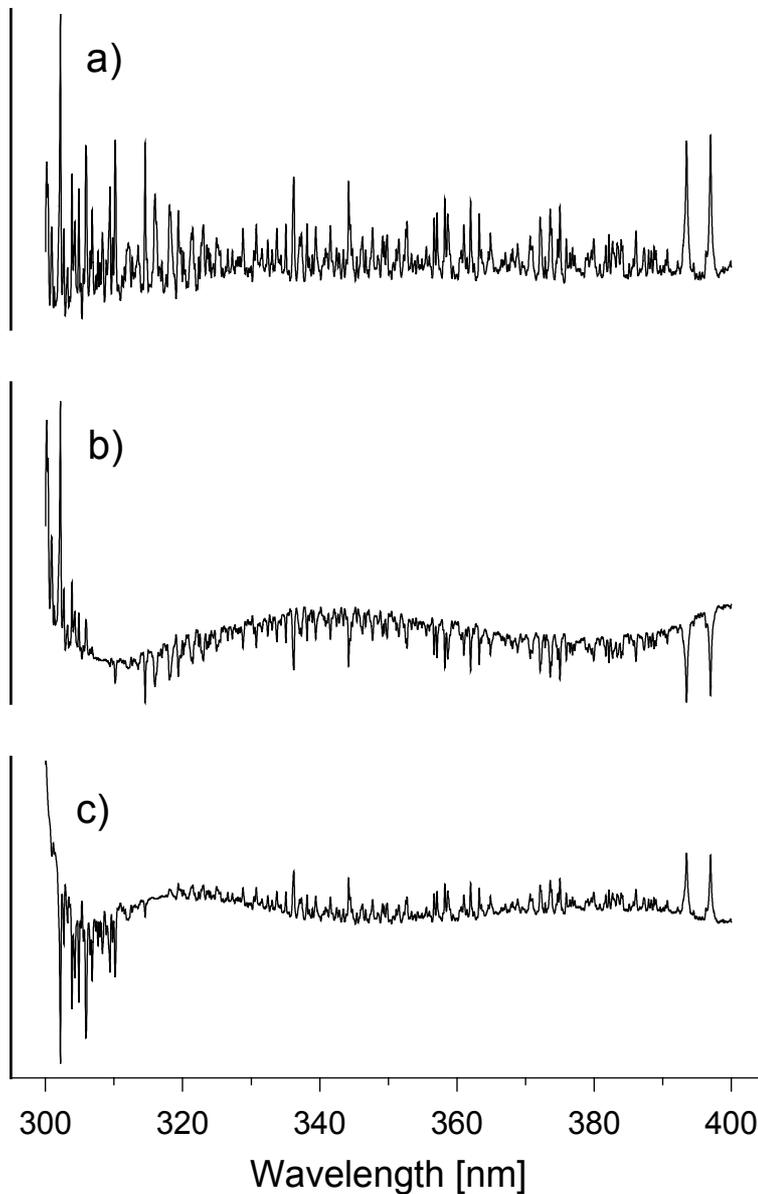


Fig. 4 Set of Ring spectra including also atmospheric absorptions. Spectra b) and c) are orthogonalized with respect to spectrum a) which was calculated without atmospheric absorptions (The calculations were performed by K. Chance).

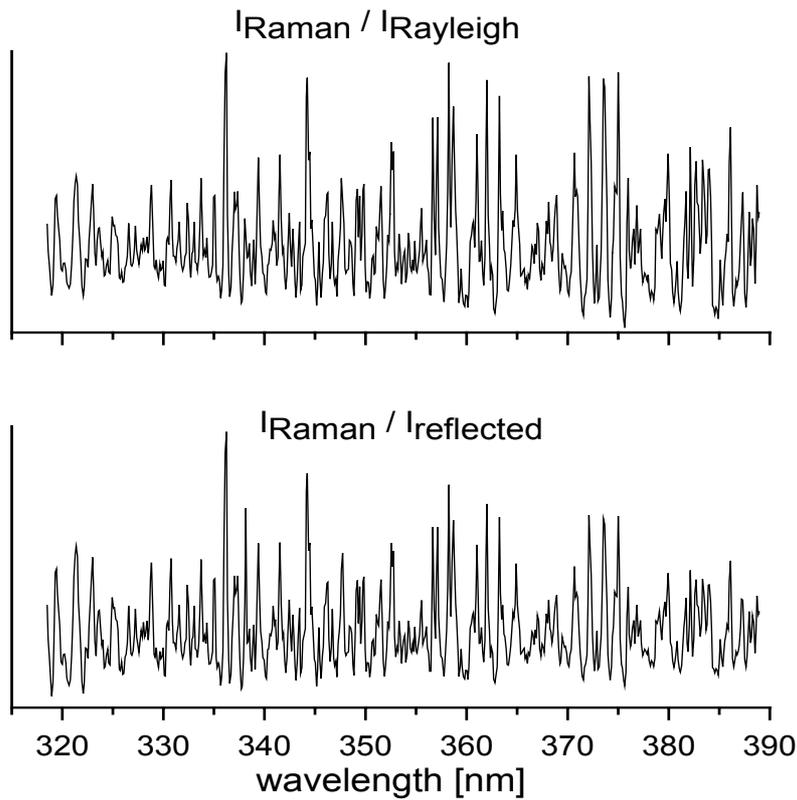


Fig. 5 Set of Ring spectra for the correction of the variation of the magnitude of the Filling-in with wavelength. The upper spectrum was calculated assuming only Rayleigh scattering. The lower spectrum was calculated assuming reflection on the (grey) earth's surface [Wagner, 1999].

## Outlook

Further improvements of the Ring correction will be possible in the future by improved atmospheric radiative transport modelling including multiple scattering and polarisation.

Standardisation of the Ring correction and recommendation for the determination of Ring spectra might be desirable in the future.

The magnitude of the Ring effect seems to be a significant indicator for cloudiness [Joiner and Bhartia, 1995; DeBeek et al., 2001]; The determination of the Ring effect for satellite observations like GOME and SCIAMACHY can therefore contribute to improved cloud correction algorithms.

## B) The $I_0$ effect

The spectral resolution of DOAS instruments in the UV/vis region is typically in the range of about a few tenths to several nanometers. Thus the natural line widths of the solar and the atmospheric absorptions are usually not spectrally resolved (see Fig. 6). The actually observed intensity  $I^*(\lambda)$  can be written as

$$I^*(\lambda) = F * I(\lambda) = \int I(\lambda') \cdot f(\lambda - \lambda') \cdot d\lambda' \quad (6)$$

Here \* indicates the convolution with the slit function of the instrument  $f(\lambda)$ . For the spectral retrieval the cross sections of the atmospheric absorbers are also convolved with this slit function.

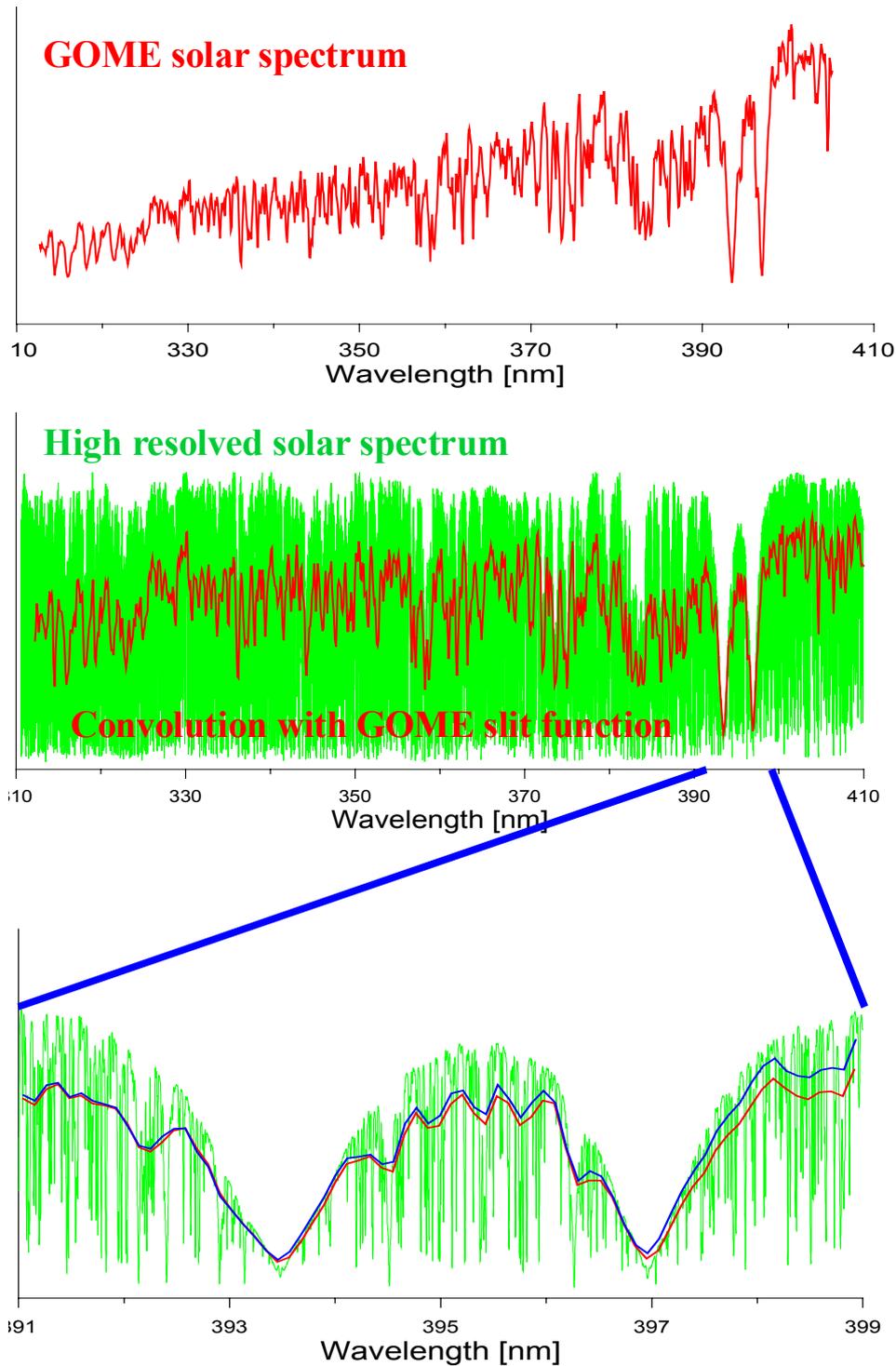


Fig. 6 Influence of the limited spectral resolution of typical DOAS instruments.  
 Top: GOME direct sun measurement (spectral resolution of about 0.2 nm FWHM).  
 Middle: High resolved solar spectrum (green) [Kurucz et al., 1981]. After convolution with a Gaussian function of about 0.2 nm FWHM the spectrum (red) becomes similar to the GOME measurement.  
 Bottom: A comparison of the convolved spectrum (blue) with the GOME measurement (red).

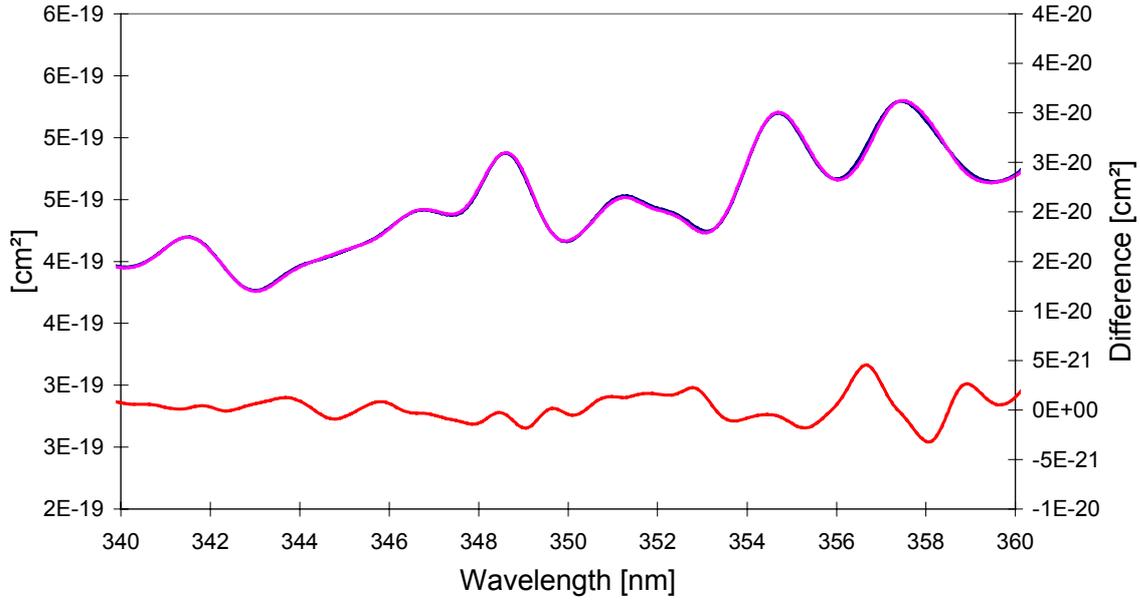


Fig. 7  $I_0$ -effect: The blue and violet curve represent  $\text{NO}_2$  absorption spectra, one spectrum was calculated for a smooth light source, the other for the highly structured solar spectrum. After convolution both spectra look nearly similar. However, the difference of both spectra shows residual structures resulting from the interference of the atmospheric absorption with the Fraunhofer lines in the solar spectrum.

Since the convolution and the logarithm can not be exchanged a non linearity between the observed optical depth  $\tau$  and the column density ( $S=\sigma \cdot \rho \cdot l$ ) of the measured trace gas results (see eq. 2). This effect which increases for strong absorbers is usually referred to as saturation effect.

In addition to this saturation effect a more subtle effect appears: an interference of the spectral absorption structures with the highly structured solar spectrum  $I_0(\lambda)$ . Dividing the measured spectrum by the Fraunhofer spectrum (eq. 2) fails in totally removing the Fraunhofer structures, because the convolution and ratioing can not be exchanged. Since this effect arises from the spectral structure of the solar spectrum  $I_0(\lambda)$  it was referred to as ‘ $I_0$  effect’ [Johnston, 1996; Platt et al., 1997, Friess, 1997; Richter, 1997; Alliwel et al., 2001]. It can be corrected for by using modified absorption cross sections which can be calculated in the following way [Johnston 1996]:

First a highly resolved solar spectrum  $I_0(\lambda)$  is convolved with the instrument function. In the next step a modelled absorption spectrum (calculated from the highly resolved solar spectrum and a highly resolved absorption cross section) is convolved with the instrument function (eq. 6). For the latter an atmospheric column density  $S$  has to be assumed. The  $I_0$  corrected cross section is then derived:

$$\sigma_{corrected}(\lambda, S) = \frac{-\ln\left(\frac{I^*(\lambda, S)}{I_0^*(\lambda)}\right)}{S} \quad (7)$$

Clearly  $I_0$ -corrected cross sections derived in this manner can perfectly match the absorptions in the measured atmospheric spectrum only if the  $S$  used for the calculation matches the atmospheric  $S$ . Usually the maximum values of typically occurring atmospheric  $S$  are assumed

for the determination of the corrected cross sections [Alliwell et al., 2001]. For an improved DOAS analysis, however, an iterative approach could be applied using the SCDs retrieved in a first fitting process to calculate  $I_0$  corrected cross sections of several absorbers. These cross sections could be used for the next DOAS fitting, and so on.

While for most of the atmospheric absorbers the  $I_0$  effect is weak and could be neglected, in some cases it has to be considered for a proper analysis. This is in particular the case when strong atmospheric absorbers like  $O_3$  have to be 'removed' in order to measure the underlying weak absorptions of other trace gases like BrO. Figure 7 shows residual structures which result from the application of a non  $I_0$ -corrected  $NO_2$  cross section for atmospheric measurements using the sun as light source.

It was shown e.g. by Alliwell et al. [2001] that for ground based measurements of BrO at mid latitudes in summer the derived BrO absorption is effected by up to 30% if the simultaneously fitted  $O_3$  cross sections are  $I_0$  corrected or not. In contrast, for BrO measurements at polar regions, in particular in winter time, the influence of the  $I_0$  effect on the BrO analysis was found to be much smaller.

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