OZONE PROFILE RETRIEVAL FROM GOME SATELLITE DATA I: ALGORITHM DESCRIPTION

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Abstract

The Global Ozone Monitoring Experiment (GOME) is a new nadir-viewing diode array spectrometer on ERS-2 detecting radiation in the UV/visible spectral range between 240 nm and 790 nm (Burrows, 1988a, Burrows, 1993). It measures the solar irradiance and the earthshine radiance. Originally designed to provide total ozone maps with global coverage the instrument performance allows to retrieve also total column information about other atmospheric species like NO_2, ClO, BrO, and SO_2. The decrease of the atmospheric optical depth between 250 nm and 350 nm enables the retrieval of height-resolved ozone information from GOME spectra, originally proposed by . The retrieval algorithm FURM (FUll Retrieval Method) has been developed at the Institute of Remote Sensing (*ife*) to admit this objective. The basis of the FURM algorithm is the forward model GOMETRAN, developed at *ife* (Rozanov, 1997). GOMETRAN calculates the radiation field of the atmosphere by considering absorption and multiple scattering processes by molecules and particles. It also simultaneously provides weighting functions to linearize the radiative transfer equation (Rozanov,1997b). A solution of the non-linear problem can be obtained by fitting the GOME spectra in an iterative process.

Keywords: GOME; Ozone Vertical Profiles

1. INTRODUCTION

As a result of the potential anthropogenic modification of atmospheric compositions, measurement of stratospheric compositions is currently of scientific and general interest. Ozone, although an atmospheric minor component has one of the strongest influences on radiation transfer in the UV/visible. Since the discovery of ozone depletion in the antarctic spring, known as the "ozone hole phenomenon" (Farman, 1985), atmospheric measurements concentrate on those components participating in the chemical and dynamical ozone processes.

Balloon-borne in situ and ground-based spectroscopic measurements are adequate to monitor well-mixed atmospheric species having relatively large lifetimes (e.g. O_2 or CO_2). However, only satellite-based monitoring instruments enable the observation of atmospheric components on a global scale. Gases such as O_3 and NO_2 are characterized by larger horizontal and vertical variability. The retrieval of ozone vertical distributions from nadir observations is possible from measurements in the ultraviolet range of the spectrum where the Hartley-Huggins bands of ozone are (Singer, 1957). This spectral range is observed by the measurements of backscattered earthshine radiation by the GOME channels 1 and 2 (237 nm to 314 nm and 311 nm to 405 nm, respectively) having a spectral resolution of 0.2 nm (Burrows, 1997).

For an evaluation of these spectra all atmospheric processes influencing the solar radiation in this spectral range have to be considered. Clearly instrumental effects on the radiation have to be taken into account. A brief description of GOMETRAN and its application for retrieval is given in chapter 2. Chapter 3 describes the theoretical background of the retrieval methods used in FURM. An overview of the retrieval algorithm is given in chapter 4. Chapter 5 contains an example of a FURM retrieval using a GOME UV/visible spectrum. Finally conclusions and an outlook are given in chapter 6.

2. THE FORWARD MODEL

An essential part of any retrieval of ozone profiles is an adequate radiative transfer model (RTM). For the simulation of GOME satellite data the RTM GOMETRAN has been developed at *ife*. GOMETRAN is based on the finite differencing method and calculates the atmospheric radiance field in spherical geometry and multiple scattering mode for the GOME spectral range (240 nm--790 nm) (Rozanov, 1997). Weighting functions (see chapter 3) are calculated simultaneously (Rozanov, 1997b). Compared to time-consuming numerical weighting function calculations the latter is a novel and powerful advance. Vertical profiles of pressure, temperature, and a number of trace gases (e.g. O_3, NO_2, ClO, and BrO) needed as input parameter for GOMETRAN are taken from a climatology calculated using the MPI-Mainz 2D-model (Brühl, 1991). Trace gas absorption cross sections are taken from different sources (for example (Richter, 1995, Dehn, 1995). The temperature dependence of ozone absorption cross sections in the Hartley-Huggins bands being accounted for (Richter, 1995).

Aerosol distributions are similar to these used in the RTM LOWTRAN/MODTRAN, the atmosphere being divided into four regimes with different aerosol types. The aerosol absorption and extinction coefficients are taken from the LOWTRAN database. The geometrical input parameters (sun-zenith angle, line-of-site angle, relative azimuth angle) are provided by the GOME level 1 data product. Measurements from the spectral broadband measurements of the three polarization measurement devices (PMD) of GOME, are used to correct for atmospheric polarization effects in the level 1 to 2 data processing. This data is also used to calculate true color pictures for cloud detection (see this paper, part II (Eichmann, 1997).

For FURM retrievals the GOME ground pixel size is 960 km x 100 km as determined by the satellite line-of-site zenith angles of +/-31^{o} for channel 1A (240 nm - 307 nm) and the integration time. The different views of the atmosphere are accounted for within the retrieval. To approximate the surface albedos for the considered viewing directions the measured PMD reflectivity is used.

The development of a GOMETRAN version for the modeling of rotational Raman scattering features in the spectrum, known as the Ring effect, is required to account for these structures (Burrows, 1996, Vountas, 1997). The parameterization of clouds in GOMETRAN has been recently finished and will be implemented in the retrieval algorithm (Kurosu, 1997).

3. PROFILE RETRIEVAL METHODS

In the ultraviolet spectral range the Hartley-Huggins bands of ozone lead to a strong absorption of the incoming solar radiation. Starting at 260 nm, where the ozone absorption is at its maximum, the penetration depth of radiation increases. From wavelengths larger than 310 nm the radiation starts to significantly reach the troposphere and beyond 340 nm the radiation reaches the surface. The maximum change of radiation due to ozone absorption moves therefore from 55 km altitude at 260 nm to about 20 km at 310 nm and larger wavelengths. Equation (1) describes the change of radiation expressed as a derivative with respect to the state vector.

$$K_{i,j} = \frac{\partial y_i}{\partial x_j} \quad ; \qquad (1)$$

where x_j is the j-th component of the atmospheric state vector x and y_i is the i-th component of y, defined as

$$\vec{y} = \ln I_{rad} - \ln I_{irr} + \ln \pi$$

I_{irr} is the incoming solar irradiance [photons.nm^{-1}.cm^{-2}.s^{-1}] and I_{rad} is the backscattered radiance [photons.nm^{-1}.cm^{-2}.s^{-1}.sr^{-1}]. The columns of the matrix K are known as weighting functions. Figure 1 shows ozone weighting functions calculated using GOMETRAN for a mid-latitude summer ozone profile. The logarithm of the sun-normalized earthshine radiance y at wavelength Lambda of an atmosphere which is in the state x can be mathematically expressed by

$$\vec{y} = F(\vec{x}) \quad ; \qquad (2)$$

F is a non-linear functional. Solving this system is called inversion. For ozone profile retrieval from backscattered UV spectra this is an under-constrained problem. To find a unique solution sufficient additional constraints have to be applied. Various sources of such information are proposed in the literature, a knowledge of the climatology being often used (see e.g. Twomey, 1977, Rodgers, 1976).

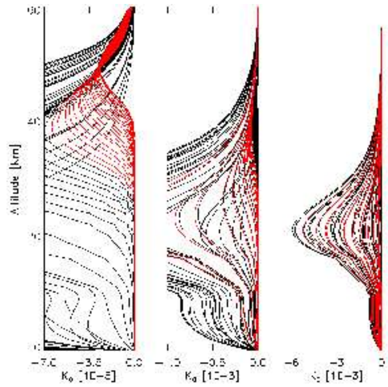

Figure 1: Ozone weighting functions. Red color for 260 nm to 285 nm (left), 289 nm to 307 nm (center), and 320 nm to 340 nm wavelengths (right) .

The first step involves solving the non-linear problem is a linearization of equation (2). The weighting function matrix K_0, calculated for the state x_0, can be used to make a linear approximation of equation (2) in the neighborhood of the linearization point x_0.

$$\vec{y} - \vec{y}_0 = K_0(\vec{x} - \vec{x}_0) \quad ; \qquad (3)$$

where y_0 is the model spectrum calculated using x_0. Expression (3) gives a set of linear equations. If e is defined as

$$\vec{e} = (\vec{y} - \vec{y}_0) - K_0(\vec{x} - \vec{x}_0)$$

the quadratic form

$$\vec{e}^T S_y^{-1} \vec{e} + d\vec{x}^T S_a^{-1} d\vec{x} = min \qquad (4)$$

has to be minimized, where dx=(x-x_a). x_a with covariance matrix S_a is used as a-priori statistical information about the atmosphere. This statistical regularization is known as the optimal estimation method (see e.g. Rodgers, 1976). Calculation of the derivative of (4) with respect to dx yields

$$\begin{aligned}\vec{x}_1 &= \vec{x}_a + (K_0^T S_y^{-1} K_0 + S_a^{-1})^{-1} K_0^T S_y^{-1} (\vec{y} - \vec{y}_0) \\ &= \vec{x}_a + D_0(\vec{y} - \vec{y}_0) \quad ;\end{aligned} \qquad (5)$$

where $D_0=(K_0^T.S_y^{-1}.K_0 + S_a^{-1})^{-1}.K_0^T.S_y^{-1}$ is known as the contribution matrix. Its columns express how much a measurement at a certain wavelength contributes to each component of the fit vector x_1. For a linear forward model the solution of equation (2) with the largest probability is obtained. The non-linear problem has to be solved iteratively following the newton iteration scheme where the fit result x_1 is used as linearization point for the next iteration. Skipping to step n+1 the calculation of the derivative as above yields

$$\vec{x}_{n+1} = \vec{x}_a + D_n((\vec{y} - \vec{y}_n) + K_n(\vec{x}_n - \vec{x}_a)) \quad ; \qquad (6)$$

where x_{n+1} is the fit profile of the (n+1)th iteration and D_n and K_n are calculated using x_n. This enables the final fit x' to be determined by iteration until a convergence is achieved. Four error sources of the fit x' can be identified, as expressed in the following equation (Rodgers, 1996):

$$
\begin{aligned}
\vec{x}' - \vec{x} &= (A - I)(\vec{x} - \vec{x}_a) && \text{(smoothing error)} \\
&\quad + DK_b(\vec{b} - \vec{b}') && \text{(model parameter error)} \\
&\quad + D\Delta f(\vec{x}, \vec{b}, \vec{c}) && \text{(forward-model error)} \\
&\quad + D\vec{\varepsilon} && \text{(retrieval noise)}
\end{aligned}
$$

where A=D.K is the model resolution matrix or averaging kernel matrix (Rodgers, 1996), b and b' are the true and estimated model parameters, respectively, c is the vector of the atmospheric parameters not considered in the model, and Epsilon is the measurement error.

Another common approach to fit the state vector to a measured spectrum is to expand it in a series of suitable basis vectors and iteratively determine the expansion coefficients. A particularly convenient base system are the Eigenvectors Psi_n of the information matrix (see Shannon, 1962), which for the (n+1)th iteration is defined as:

$$P = S_a K_n^T S_y^{-1} K_n$$

If we define $Alpha_{n,i}$ as

$$\vec{\alpha}_{n,i} = K_n^T S_y^{-1} K_n \vec{\psi}_{n,i}$$

the basis systems Psi_n and $Alpha_n$ are orthogonal:

$$\vec{\psi}_{n,i}^T \vec{\alpha}_{n,j} = \delta_{i,j} \nu_i$$

Now x_{n+1} can be written as a linear combination of the Eigenvectors Psi_n:

$$\vec{x}_{n+1} = \vec{x}_a + \sum_i^{n_e} \beta_{n+1,i} \vec{\psi}_i;$$

with

$$\beta_{n+1,i} = \frac{\lambda_i}{\nu_i(1+\lambda_i)} \vec{\psi}_i^T K_n^T S_y^{-1} \left[\vec{y} - \vec{y}_n + K_n(\vec{x}_n - \vec{x}_a)\right];$$

where $Lambda_i$ is the i-th Eigenvalue of P. The information content of a measurement can be defined as the reduction in entropy of the ensemble of possible states due to the measurement. In this case the Eigenvectors of P correspond to the independent pieces of information contained in the measurement (Kozlov, 1983). Both the optimal estimation and the Eigenvector method are implemented in the FURM algorithm.

4. THE FURM ALGORITHM

The profile fit parameters used in the retrieval process are ozone number densities at 61 equidistant altitude levels between 0 km and 60 km or an adequate number of Eigenvector coefficients. Fitted as scalar parameters are first, the Rayleigh scattering coefficients and second, the aerosol extinction coefficients, both integrated over the atmosphere. The third scalar fit parameter is the surface albedo. Furthermore the temperature used for the calculation of temperature dependent ozone absorption cross sections in the Hartley-Huggins bands is fitted. The NO_2 total column is a useful fit parameter in GOME channel 2. A correction of the spectral structures due to the Ring effect is essential. Therefore a database of synthetic Ring spectra calculated for 11 different sun zenith angles has been prepared (Vountas, 1997). These spectra are used as Ring weighting functions. Thus the Ring parameter expresses the relative Ring amplitude change.

The described fit parameters represent the components of the atmospheric state vector x. All other atmospheric parameters are treated separately as constant input for the forward model. Climatology data derived by the MPI-Mainz 2D-model is taken to be the a-priori mean state x_a. x_a is also used as first linearization point x_0 (first guess).

The a-priori covariance matrix is calculated as

$$S_{a_{i,j}} = \sigma^2 \exp^{-|z_i - z_j|/r};$$

where Sigma is the standard deviation, z_i and z_j are altitudes, and r is the correlation radius. Except for the Ring amplitude parameter and the temperature all weighting functions are prepared for the fit parameters to be the relative differences or correction factors.

For measurements of the radiance, irradiance and absorption cross section spectra shifts and/or deformations (squeezes) will be present. This is due to thermally induced deformations of the GOME instrument which cause dislocation of the detector array. A shift-and-squeeze module for the FURM was developed at *ife* to correct for the residual misregistration of the wavelength.

Figure 2 shows a schematic overview of the main algorithm modules. After reading the measurement spectrum evaluated and setting up the a-priori statistic as described GOMETRAN calculates the model spectrum with the first-guess. Before applying the estimation formula the model, irradiance, and Ring spectra have to be ``shifted and squeezed'' with respect to the measured radiance. The fit result is then used to setup a new input data set for the calculation of the next model spectrum. A convergence test is carried out to decide whether to start the next iteration or to stop after a quality check.

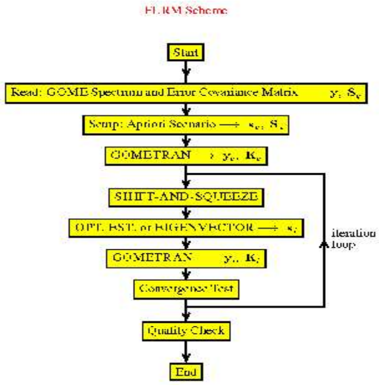

Figure 2: Basic structure of FURM

Convergence is tested by comparing the number of spectral points m with the chi^2 value of the actual and previous model spectra.

$$\chi^2 = (\vec{y}_{n-1} - \vec{y}_n)^T S_y{}^{-1} (\vec{y}_{n-1} - \vec{y}_n) \ll m$$

if m is an order of magnitude smaller than chi^2 convergence has occurred.

Another appropriate convergence test is to compare the last two fit results as follows:

$$d^2 = (\vec{x}_n - \vec{x}_{n+1})^T S^{-1} (\vec{x}_n - \vec{x}_{n+1}) \ll n_p \quad ;$$

where n_p is the number of fit parameters. If in the n-th iteration convergence has occurred, S, the covariance matrix of the retrieval error (smoothing error plus retrieval noise), is given by

$$S = (K_n^T S_y^{-1} K_n + S_a^{-1})^{-1}$$

After convergence has occurred a quality check is performed which decides whether the fit result corresponds to the measurement spectrum within measurement error, thus provided the condition

$$\chi^2 = (\vec{y} - \vec{y}_{n+1})^T S_y^{-1} (\vec{y} - \vec{y}_{n+1}) \approx m$$

5. APPLICATION ON GOME SPECTRA

A systematic evaluation of GOME spectra using the retrieval algorithm FURM is being achieved as support of this years European arctic winter campaign providing near-real-time ozone maps and selected vertical ozone profiles within 24-48 hours. For a description of this activities see (Eichmann, 1997). This chapter gives an example of a FURM retrieval of GOME spectra with both the Eigenvector and the optimal estimation methods.

For the retrieval of GOME spectra the scanning scheme of the instrument has to be taken into account whether to assure a consistent measurement in both channel 1A and channel 2 or if not all channel 2 spectra are used to make an adequate scanning simulation. The integration time of a channel 2 spectrum is 1.5 s, which leads to an observed area of 320 x 40 km (GOME ground-pixel) whereas the channel 1A integration time of 12 s yields about 960 x 100 km. For the following example eight channel 2 spectra were integrated corresponding to one spectrum in channel 1A.

Figure 3 shows the fit results obtained from a GOME spectrum measured at the 4th of April 1996 (ground-pixel center: 52.7 N, 2.4 E). For comparison an independent ozone profile measurement from a sonde observation at Debilt (52.1 N, 5.2 E) on the same day was available.

Both, the optimal estimation and the Eigenvector method provide ozone number densities for 61 altitude levels. To find an adequate profile form for presenting more or less independent quantities the averaging kernels (see chapter 3) of a typical retrieval case have been analyzed. As the averaging kernels express the response of the fit vector to a delta function perturbation in the true state its full-width-at-half-maximum for example can lead to a rough approximation of what is called vertical resolution. Thus as first approach for a reasonable devision the profiles were integrated for 7 layers of thicknesses as shown. This devision was also applied on the sonde data. Above the largest used sonde height (z_{max}=33 km) the sonde profile was expanded to 60 km by using the climatology ozone profile corrupted by a suitable factor to avoid a jump. This or similar assumptions (e.g. a constant volume-mixing-ratio) have to be made when ozone total column amounts are to be determined from sonde ozone profiles which have to be considered when comparing the total ozone amounts (see Figure 3). Corresponding layer contents as for the fit results were also calculated for the sonde profile and for the a-priori profile to have adequate quantities for a comparison. To determine the a-priori covariance a standard deviation of 30 % and a correlation radius of 5.2 km were chosen. The retrieval have been achieved for the spectral window from 289 to 307nm (channel 1A) and for 320 to 340 nm (channel 2). For the optimal estimation retrieval only the channel 1A window was chosen as a successful quality check could not yet obtained using this method in combination with a multi-window retrieval. This can probably be explained by inconsistencies of channel 1A and channel 2 modeling due to forward-model parameter errors which are not yet fully analyzed. Anyway, a multi-window retrieval can be achieved by using the Eigenvector method using naturally that number of fit parameters which correspond to the information content of the observing system. Therefore this method is not susceptible to instabilities.

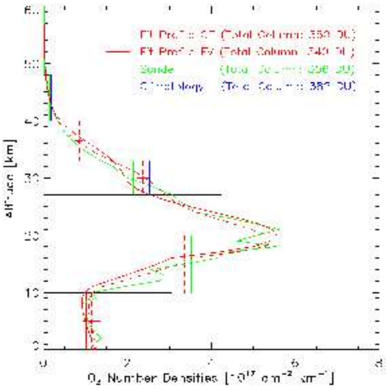

Figure 3: GOME ozone profile from April 4 1996 (52.7 N, 2.4 E) versus sonde profile (Debilt, 52.1 N, 5.2 E). Dashed red line: fit using a channel 1A spectrum (289-307nm) with the optimal estimation method. Solid red line: multi-window fit (289-307nm and 320-340 nm) using the Eigenvector method. .

For the considered case (see Figure 3) both methods lead to quite good agreement with the Debilt-sonde. Above 20 km the result of the Eigenvector method is more located to the sonde whereas below 20 km the optimal estimation result shows better agreement. This is not a surprise when considering that due to the lack of tropospheric information in channel 1A the optimal estimation retrieval tends to the a-priori which is near the sonde profile below 20 km. Retrievals with low ozone scenarios have also shown this behavior.

Figure 4 shows the spectral residuum between the measured spectrum and the model spectrum obtained from the fit result using the Eigenvector method expressed as difference of the logarithms of the sun-normalized earthshine radiances. It is straightforward to show that this is for small differences also a measure of the relative spectral difference. The maximum difference is about 0.7 % and the mean difference over the whole spectral range is lower than 0.2 %. For the optimal estimation method almost the same values were obtained for channel 1A. Both results are far below the measurement error boundaries to what was aspired by the FURM quality check.

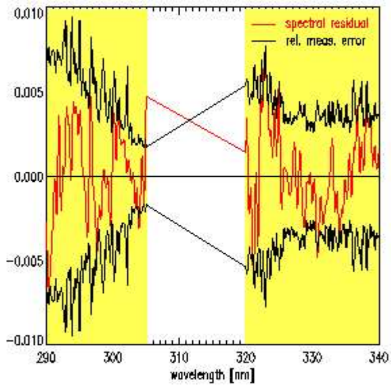

Figure 4: Spectral residuum for the retrieval using the Eigenvector method (see Fig. 5). .

More examples of comparisons between sonde ozone profile measurements and FURM retrieval results are given in (Eichmann, 1997). A more detailed validation of FURM will be achieved in the frame of Phase II of the European arctic winter campaign activities at *ife*.

6. CONCLUSIONS AND OUTLOOK

Both the optimal estimation and the Eigenvector method proved to be appropriate for the retrieval of height resolved ozone information from GOME data.
However, the Eigenvector method is more flexible in that the number of Eigenvectors can be varied according to stability criteria imposed on the solution.

Studies based on synthetic measurements show that ozone absorption in the Hartley bands allows the retrieval of stratospheric ozone information from channel 1a.
Taking into account the temperature dependent ozone absorption in the Huggins band by simultaneously fitting 1a and 2b-spectra extends the range of height-resolved information downwards into the troposphere.
In this case an appropriate cloud parameterization as well as adequate surface albedo and aerosol parameters are crucial.
The inclusion of a temperature fit parameter can improve the tropospheric fit results.

First results with real GOME spectra show that the spectral residuals after the fit are within the range of measurement errors.
Total ozone values derived with DOAS (operational product) and FURM show good agreement for a variety of atmospheric conditions.
FURM is able to retrieve low ozone scenarios even if the a-priori profile is considerably higher.

So far, O_3-profile retrievals using channel 2 are restricted to cloud-free scenarios. However, in the near future an already existing GOMETRAN cloud parameterization (see Kurosu, 1997) will be implemented in FURM.

Furthermore, it has to be investigated if additional fit windows in the Chappuis bands (channels 3 and 4) can improve the fit results.

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