The need of polarization modeling for ozone profile retrieval from backscattered sunlight

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[1] In the ultraviolet and visible part of the spectrum, measurements of space-borne grating spectrometers are in general sensitive to the state of polarization of the observed light. The correction for this polarization sensitivity is based on broadband polarization measurements. In parts of the spectrum where the state of polarization is varying rapidly with wavelength this correction is not sufficient and severely limits the accuracy of the atmospheric parameters retrieved from the polarization corrected measurements. In this paper we demonstrate that the problems due to instrument polarization sensitivity can be solved in a natural way by the use of polarization modeling. For the forward model of a retrieval algorithm we propose the combination of a vector radiative transfer model to simulate the transport of radiation in the probed atmosphere and a straightforward simulation of the instrument polarization sensitivity by use of the Mueller matrix formalism. The use of a vector radiative transfer model also overcomes another common bias in retrieval algorithms, caused by the widely used scalar approximation of atmospheric radiative transfer. The capability and need of the proposed approach are demonstrated for ozone profile retrieval from measurements of the Global Ozone Monitoring Experiment (GOME). A comparison of retrieved profiles with 123 ozonesonde profiles shows that the use of a polarization forward model yields a significant improvement in root-mean square difference of about a factor 1.5 in the stratosphere as well as in the troposphere. Also, a solar zenith angle dependence in the differences is reduced significantly. *INDEX TERMS:* 1610 Global Change: Atmosphere (0315, 0325); 1694 Global Change: Instruments and techniques; 1640 Global Change: Remote sensing; 3210 Mathematical Geophysics: Modeling

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1. Introduction

[2] Satellite measurements of backscattered sunlight contain essential information about the global distribution of atmospheric constituents. For example, the backscattered ultraviolet (BUV) and solar backscattered ultraviolet (SBUV/2) instruments have shown to provide height-resolved information about ozone. These instruments measure the backscattered ultraviolet radiances at 12 wavelengths between 252 and 340 nm, allowing the retrieval of ozone profiles above $\sim$25 km. The Global Ozone Monitoring Experiment (GOME) launched in 1995 on board of the second European Remote Sensing satellite (ERS-2), performs measurements at $\sim$0.2 nm resolution in the spectral range 240–800 nm in nadir viewing geometry. The fine wavelength resolution of GOME combined with a high signal-to-noise ratio implies the potential for ozone profile retrieval below 25 km [*Chance et al.*, 1997; *Munro et al.*, 1998; *Hoogen et al.*, 1999; *Hasekamp and Landgraf*, 2001]. For information about the GOME instrument see the overview paper of *Burrows et al.* [1999]. In March 2002 the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) has been launched on ESA's ENVISAT-1 satellite. This instrument has an extended spectral range 240–2880 nm and will also perform measurements in limb and occultation geometry, in addition to nadir measurements. In the near future, several GOME-type instruments are scheduled for launch. The Ozone Monitoring Instrument (OMI), to be launched in 2003 on NASA's EOS-AURA satellite will measure in the spectral range 270–500 nm at about 0.5 nm resolution. Additionally three GOME-2 instruments will be flown on the METOP series of EUMETSAT, starting with METOP-1, scheduled for launch in 2005.

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[3] Each retrieval of atmospheric constituents requires a forward model **F** that describes how the measurement vector **y** depends on the atmospheric state vector **x**,

$$\mathbf{y} = \mathbf{F}(\mathbf{x}) + \mathbf{e}, \tag{1}$$

with error vector **e**. Here, the forward model **F** simulates the transfer of radiation through both the atmosphere and the instrument. The elements of the *N*-dimensional atmospheric state vector **x** are the atmospheric parameters to be retrieved, for example, averaged trace gas densities in different altitude layers of the atmosphere, aerosol or cloud parameters. Each element of the *M*-dimensional vector **y** corresponds to a single measurement, for example, a radiance measurement at a certain wavelength. The inversion of (1) yields the unknown atmospheric state vector **x**. Obviously, the quality of the retrieved atmospheric parameters depends strongly on the accuracy of the measurements and forward model, respectively.

[4] Light reflected from the Earth's atmosphere is polarized because of scattering of unpolarized sunlight by air molecules and aerosols. A widely used approach in forward models that intend to interpret radiance measurements is to employ the scalar approximation for atmospheric radiative transfer, which neglects any polarization properties of radiation. However, it has already been pointed out by *Chandrasekhar* [1960] that due to multiple scattering of sunlight by molecules, the error of a radiative transfer model that neglects polarization can be as large as 10%, depending mainly on the viewing scenario. So, the employment of a scalar radiative transfer model in retrieval algorithms may limit the accuracy of the retrieved atmospheric parameters. *Mishchenko et al.* [1994] gave an extensive overview of the dependence of the radiance error due to neglecting polarization, on various parameters of a homogeneous molecular atmosphere, while *Lacis et al.* [1998] discussed the same problem for aerosol loaded atmospheres. *Stammes* [1994] evaluated the scalar error for a typical GOME nadir-viewing scenario.

[5] Also, the transport of radiation through the instrument cannot be simplified by a scalar approximation. Here, the different optical devices in the instrument are normally sensitive to the state of polarization of light. The interaction of polarized light with polarization-sensitive optical devices yields a different radiance that is measured by the detectors than the radiance that enters the instrument. In this paper we refer to the radiance that is measured by the detectors as the polarization-sensitive measurement.

[6] In the OMI and the SBUV/2 instruments the problem of instrument polarization sensitivity is avoided because the polarized backscattered sunlight is depolarized before it interacts with the polarization-sensitive optical components. Obviously, for these instruments it is still necessary to take polarization into account in the forward model in order to model the radiance correctly.

[7] For GOME, SCIAMACHY, and GOME-2 it is intended to eliminate the polarization response of the instrument from the polarization-sensitive measurement. This part of the data processing is called the polarization correction. For the polarization correction the state of polarization of the backscattered sunlight is estimated from broadband polarization measurements, performed by Polarization Measuring Devices (PMDs).

[8] For GOME the polarization correction is based on only three broadband PMDs, which have turned out to be not sufficient in regions of the spectrum where the state of polarization changes rapidly with wavelength [*Aben et al.*, 1999], like in the Oxygen-A band [*Stam et al.*, 2001] and the Hartley and Huggins ozone absorption bands [*Schutgens and Stammes*, 2002]. Here, the polarization correction limits the accuracy of the atmospheric parameters that are retrieved from the radiance measurements. The PMDs of SCIAMACHY have bandwidths comparable to those of GOME, so similar problems are expected for the polarization correction of this instrument. For GOME-2 a much better spectral coverage will be obtained for the polarization measurements in most parts of the spectrum but not in the Oxygen-A band region. In general, the current treatment of instrument polarization sensitivity for GOME, SCIAMACHY, and GOME-2 causes an intrinsic error in the calibrated radiances. Apart from the lack of information about the spectral dependence of polarization, also the quality of the broadband polarization measurement may limit the accuracy of the polarization correction. It has been shown by *Tanzi et al.* [1999] that the performance of the PMD of GOME in the range 300–400 nm is changing severely in time, which obviously enlarges the polarization correction error.

[9] In this paper we show that a proper way to treat the polarization sensitivity of the instrument is to use a forward model that employs the complete vector approach for radiative transport through the atmosphere as well as through the instrument. For this purpose retrievals have to be performed on non-polarization-corrected radiances. The capability and need of this approach is demonstrated for ozone profile retrieval from GOME measurements. In section 2 we describe measurement principles of a polarization-sensitive instrument and discuss the concept of polarization correction. In section 3 we evaluate the errors that are caused by using polarization corrected radiances and a scalar radiative transfer model, for ozone profile retrieval from GOME. The simulation of polarization-sensitive measurements by a forward model is discussed in section 4, and the advantage of this method is demonstrated by a validation of ozone profiles retrieved from GOME with 123 collocated ozonesonde measurements.

2. Measurements of a Polarization-Sensitive Instrument

2.1. Mueller Matrix and Polarization Sensitivity

[10] The radiance and state of polarization of light at a certain wavelength can be described by an intensity vector **I** which has the Stokes parameters *I*, *Q*, *U*, and *V* as its components [*Chandrasekhar*, 1960; *Hansen and Travis*, 1974]:

$$\mathbf{I} = [I, Q, U, V]^T, \tag{2}$$

where *T* indicates the transposed vector. In this paper the intensity vector is defined with respect to the local meridian plane as a reference plane.

[11] Although the aim of grating spectrometers like GOME, GOME-2, and SCIAMACHY is to measure the first component of the intensity vector, that is, the radiance *I*, they

are also sensitive to the state of polarization of the backscattered sunlight. The change in intensity vector due to interaction of the light with the different optical devices in the instrument can at a certain wavelength λ be described by

$$\mathbf{I}'(\lambda) = \mathbf{M}(\lambda)\,\mathbf{I}(\lambda), \tag{3}$$

where $\mathbf{I}$ is the intensity vector of the light that has entered the instrument, $\mathbf{I}'$ is the intensity vector of the light that eventually illuminates the detector pixels, and $\mathbf{M}$ is the instruments 4×4 Mueller matrix [see, e.g., *Coulson*, 1988]. The radiance I_{det} that is measured by a certain detector pixel can be written as

$$I_{\text{det}} = \int_0^\infty d\lambda\, \phi(\lambda) \int_0^\infty d\lambda'\, s(\lambda, \lambda')\, I'(\lambda'), \tag{4}$$

where the integration over λ' represents the effect of the instrument slit characterized by a slit function s, and the integration over λ represents the sampling of the detector pixel with a normalized sensitivity ϕ. For each detector pixel an effective Mueller matrix $\bar{\mathbf{M}}$ may be defined such that (4) can be written as

$$I_{\text{det}} = \bar{M}_{11}\, I_{\text{i}} + \bar{M}_{12}\, Q_{\text{i}} + \bar{M}_{13}\, U_{\text{i}} + \bar{M}_{14}\, V_{\text{i}}, \tag{5}$$

where I_{i}, Q_{i}, U_{i}, and V_{i} are the components of the intensity vector

$$\mathbf{I}_{\text{i}} = \int_0^\infty d\lambda\, \phi\lambda(\lambda) \int_0^\infty d\lambda'\, s(\lambda, \lambda')\, \mathbf{I}(\lambda'). \tag{6}$$

In the remainder of this paper we will express $\bar{\mathbf{M}}$ as $\mathbf{M}$, and refer to it as the Mueller matrix.

2.2. Conversion to Calibrated Radiances

[12] The aim of a calibration process normally is to convert the detected radiance I_{det} to the radiance I_{i}. This conversion can be described in two steps. The first step accounts for the radiance response of the instrument and yields the polarization-sensitive measurement I_{pol},

$$I_{\text{pol}} = \frac{1}{M_{11}}\, I_{\text{det}} = I_{\text{i}} + m_{12} Q_{\text{i}} + m_{13} U_{\text{i}} + m_{14} V_{\text{i}}, \tag{7}$$

where $m_{12}, \ldots, m_{14}$ are the relative elements of the Mueller matrix $M_{12}/M_{11}, \ldots, M_{14}/M_{11}$. This first calibration step only depends on characteristics of the instrument.

[13] The second calibration step is the conversion of I_{pol} to I_{i}. This conversion, referred to as the polarization correction, can be written as

$$I_{\text{i}} = C_{\text{pol}}\, I_{\text{pol}} \tag{8}$$

with the conversion factor

$$C_{\text{pol}} = \frac{1}{1 + m_{12}\, q + m_{13}\, u + m_{14}\, v}. \tag{9}$$

Here, q, u, and v are the relative Stokes parameters $\frac{Q_{\text{i}}}{I_{\text{i}}}$, $\frac{U_{\text{i}}}{I_{\text{i}}}$, and $\frac{V_{\text{i}}}{I_{\text{i}}}$.

[14] For the purpose of the polarization correction in (8) the elements $m_{12}, \ldots, m_{14}$ should be determined before launch in an environment that is representative for the in-flight situation. Furthermore, the relative Stokes parameters q, u, and v of the backscattered sunlight need to be known, which requires additional polarization measurements. However, in general v is significantly smaller than the other Stokes parameters [*Hansen and Travis*, 1974], so its contribution in (9) can be neglected. So, q and u have to be known at the same spectral resolution as the measurement I_{pol} is made. However, for GOME, SCIAMACHY, and GOME-2, polarization measurements are made on a much coarser spectral resolution. For example, GOME measures I_{pol} with a spectral resolution of 0.2–0.4 nm whereas the polarization measurements have a spectral resolution of 100–200 nm. Thus, the polarization spectrum in the higher spectral resolution has to be reconstructed on basis of the broadband polarization measurements. Such an approach introduces significant errors in the radiances, in parts of the spectrum with strong absorption bands. In particular, this is the case for the spectral range 300–330 nm, which is of vital importance for ozone profile retrieval [*Chance et al.*, 1997]. Here, the strong decrease of ozone absorption allows an increase in multiple scattering and causes a strong variability in the polarization properties of the backscattered sunlight [*Aben et al.*, 1999; *Oikarinen*, 2001]. This polarization feature of the backscattered sunlight cannot be reproduced by the broadband polarization measurements, which causes problems for ozone profile retrieval [*Spurr*, 2001]. The error in polarization corrected radiances of GOME in the spectral range 290–340 nm and its effect on ozone profile retrieval will be investigated in section 3. These errors are also representative for SCIAMACHY, which will perform similar broadband polarization measurements. GOME-2 will perform polarization measurements in spectral bands which are much narrower, and is thus better able to reproduce polarization structures as mentioned above. Nevertheless, the polarization measurements are not performed at the same spectral resolution as the radiance measurement so an intrinsic uncertainty due to the polarization correction remains.

3. Effect on Ozone Profile Retrieval From GOME

[15] For the calculations in this section we employ the inversion model described by *Hasekamp and Landgraf* [2001] using Phillips-Tikhonov regularization [*Phillips*, 1962; *Tikhonov*, 1963; *Hansen and O'Leary*, 1993]. In Appendix A of this paper an overview is given of this inversion method. The effect of polarization dependent errors on the retrieved ozone profile is calculated according to the formalism of *Rodgers* [1990]. For the retrieval we employ the spectral range 290–315 nm together with the window 335–340 nm. The latter window is needed to include in order to fit the surface albedo.

3.1. GOME Polarization Sensitivity

[16] For GOME it is assumed that the instrument is only polarization sensitive with respect to the Stokes parameter Q, that is, the instrument has a different sensitivity to light polarized parallel to the reference plane than to light polarized perpendicular to the reference plane (note that

the instrument's optical plane and the local meridian plane coincide). For the radiance I_{det} (5) that is measured by the detector pixels we can write

$$I_{det} = a_l I_l + a_r I_r, \quad (10)$$

where I_l and I_r are the components of the light reflected by the Earth atmosphere polarized parallel and perpendicular to the reference plane, and a_l and a_r are the sensitivities of the instrument to the two different components. In (10) I_l and I_r are already slit averaged and sampled over the detector pixel as in (6).

[17] Given the definition of I and Q [*Chandrasekhar*, 1960]

$$I = I_l + I_r \quad (11)$$

$$Q = I_l - I_r, \quad (12)$$

we can express the polarization-sensitive measurement I_{pol} (7) as

$$I_{pol} = I_i + \frac{1-\eta}{1+\eta} Q_i, \quad (13)$$

where η is the relative polarization sensitivity

$$\eta = \frac{a_r}{a_l}. \quad (14)$$

It is obtained from pre-flight calibration on the spectral resolution on which I_{pol} is measured. So, for the relative elements of the Mueller matrix (7) we find for GOME: $m_{12} = \frac{1-\eta}{1+\eta}$, $m_{13} = 0$, and $m_{14} = 0$.

3.2. Polarization Correction Error

[18] For the polarization correction (8) of the GOME instrument, the relative Stokes parameter q has to be known at the same resolution as I_{pol}, thus at about 0.2 nm. Figure 1 shows a spectrum of the relative stokes parameter q in the range 290–340 nm. Below a certain wavelength boundary at around 300 nm the main part of the measured light has scattered only once, because the strong ozone absorption makes the probability of multiple scattering very small. Assuming pure Rayleigh scattering, q depends only on the solar and viewing geometry, for singly scattered light. Thus below about 300 nm q can be calculated without any knowledge about the observed atmosphere. At longer wavelengths multiple scattering starts to influence the radiation transport and q gets clearly affected by the state of the atmosphere. To reconstruct the dependence of q on wavelength, GOME performs only three polarization measurements in the bands 300–400 nm, 400–600 nm, and 600–800 nm. Based on these three measurements and the single scattering calculation q is reconstructed by an interpolation scheme. In this paper we refer to the interpolation scheme as used by the official GOME Data Processor (GDP) for measurements made before June 1998. After that date the readout of the detectors in the UV changed which causes a slightly different interpolation scheme, which is not discussed here. For an error discussion related to the interpolation scheme after June 1998, see *Spurr* [2001] and *Schutgens and Stammes* [2002].

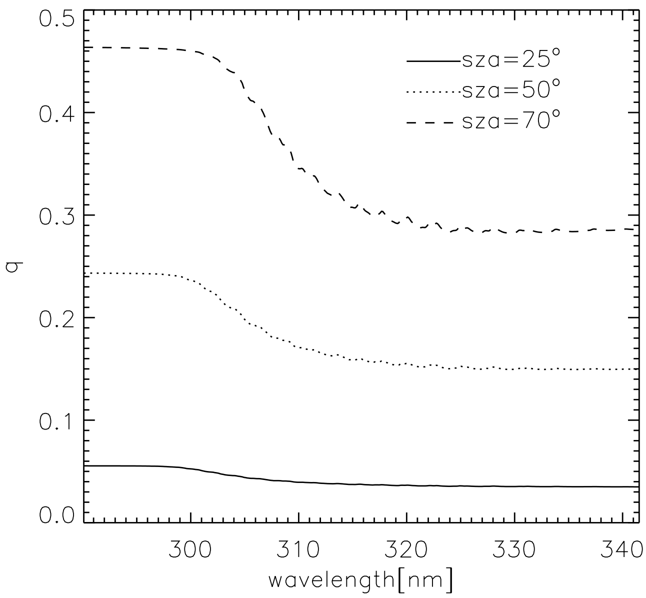

Figure 1. Relative Stokes parameter q as a function of wavelength for different solar zenith angles (SZA). The calculations are done for a Rayleigh scattering atmosphere, an ozone profile taken from an ozonesonde, and a surface albedo of 0.1. The viewing angle is integrated over a typical GOME ground pixel of size 960 × 80 km^2. The edges of the GOME ground pixel correspond to a viewing zenith angle of ±30°. The relative azimuth angle varies from 65° to −115°.

[19] In the interpolation scheme, q is calculated for wavelengths <307 nm in single scattering approximation, whereas in the spectral range 307–340 nm q is approximated by a generalized distribution function (GDF), which yields

$$q_{gdf} = \bar{p} + \frac{w_o\, e^{-(\lambda-\lambda_o)\,\beta}}{[1 + e^{-(\lambda-\lambda_o)\,\beta}]^2}, \quad (15)$$

where λ denotes wavelength. The parameters $\bar{p}$, w_o, λ_o, and β are found from the polarization measurements and from the single scattering calculation [*Balzer et al.*, 1996]. The GOME polarization correction contains errors, partly caused by the interpolation scheme [*Schutgens and Stammes*, 2002], and partly caused by the fact that the single scattering value of q is used up to 307 nm. From Figure 1 it is obvious that the single scattering approximation introduces significant errors in the range 300–307 nm, and that the interpolation causes errors for wavelengths >307 nm.

[20] The left panel of Figure 2 shows the overall error in the spectrum between 290 and 340 nm, due to the polarization correction approach mentioned above for an albedo of 0.8, and for different values of the solar zenith angle. The effect of this error on the retrieved ozone profiles is shown in the right panel of Figure 2. A clear feature is the increase of radiance and profile error with solar zenith angle. The reason for this is that the relative Stokes parameter q of the

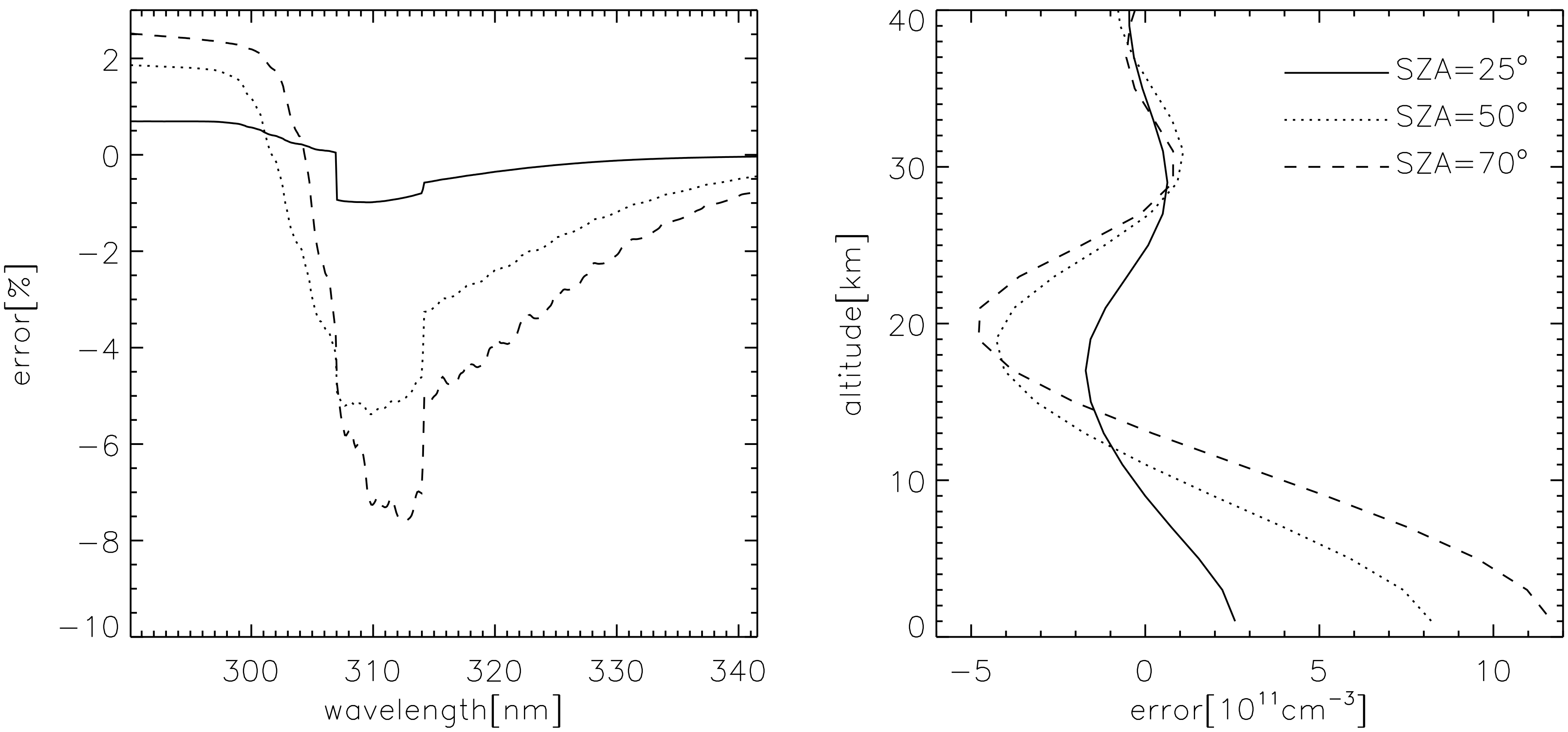

Figure 2. (left) Relative error in GOME radiances due to polarization correction and (right) effect of this error on retrieved ozone profiles. Same model atmosphere and viewing scenario as in Figure 3 but for a surface albedo of 0.8. The corresponding ozone profile is depicted in the caption of Figure 3. For the calculations a Gaussian measurement noise is assumed that is representative for GOME.

backscattered sunlight depends strongly on the viewing scenario. For the viewing geometry of GOME a small solar zenith angle yields backscattered sunlight that is less polarized than backscattered sunlight corresponding to a large solar zenith angle. An increase in multiple scattering causes a decrease in the absolute value of q, so if $|q|$ is already small for single scattered light the changes in q at $\sim$300 nm due to multiple scattering are small. This means that the error due to the use of the single scattering calculation up to 307 nm as well as the interpolation errors in the range 307–340 nm, increase with solar zenith angle. It is important to note that the above mentioned errors decrease when the surface albedo decreases. Additional to the errors discussed above also an error is present in the polarization correction due to the integration over all viewing angles corresponding to a certain ground pixel. In fact, the polarization corrected radiance should be integrated over viewing angle. However, due to the integrated readout of the instrument I_{pol} and C_{pol} (see equation (8)) are integrated separately over viewing angle in the GOME Data Processor. This integration approach causes the offset in Figure 2.

[21] For a proper interpretation of the ozone profile errors in Figure 2, the corresponding ozone profile as well as the retrieval noise [*Rodgers*, 2000] are depicted in Figure 3. From Figures 2 and 3 it follows that the maximum profile error caused by the polarization correction can be as large as about 12% in the stratosphere and 200% in the troposphere (solar zenith angle = 70°, albedo = 0.8). For all cases, the error is significantly larger than the retrieval noise.

3.3. Scalar Approximation for Radiative Transfer

[22] As shown in section 3.2, the conversion of the polarization-sensitive radiance I_{pol} to the radiance I_{i} in (6), yields significant errors in the radiances and ozone profiles. The idea behind the polarization correction is to obtain a calibrated radiance that does not depend on the polarization properties of the backscattered sunlight. For retrieval of atmospheric parameters from polarization corrected radiances, the forward model (1) only needs to simulate the radiance of the backscattered sunlight. For this purpose, it is common use to employ the scalar approximation for atmospheric radiative transfer, which neglects all polarization properties of light [*Rozanov et al.*, 1997; *Landgraf et al.*, 2001; *Spurr et al.*, 2001]. The use of the scalar approximation of radiative transfer greatly simplifies the calculations and is computationally much less expensive than the vector approach. However, for multiple scattering radiative transfer calculations, polarization have to be taken into account in order to model the radiance correctly. The neglect of polarization introduces errors in the modeled radiances which can be as large as 10% [*Mishchenko et al.*, 1994; *Stammes*, 1994; *Lacis et al.*, 1998]. Here, we shortly summarize the importance of the scalar radiative transfer error for ozone profile retrieval.

[23] The left panel of Figure 4 shows the error of a scalar atmospheric radiative transfer model, for the same viewing scenario as used in Figure 2. Here a surface albedo of 0.1 is chosen. Note that the scalar error decreases with increasing albedo. Below $\sim$300 nm the error is very small, because in this spectral region mainly single scattering takes place. For single scattered light the scalar approximation yields the same radiance as the vector approach, because the incoming sunlight can be assumed unpolarized [*Hansen and Travis*, 1974]. However, if a second scattering process takes place, which is very likely for wavelengths $>\sim$300 nm, the incoming light for this scattering process is strongly polarized [see, e.g., *Mishchenko et al.*, 1994]. The radiance of this second order scattered light does not only depend on the radiance of the singly scattered light, but also on the Stokes parameters Q and U. Thus, a neglect of polarization would lead to an incorrect value of the modeled radiance. The same is true for higher order scattering but here the effect is smaller. For wavelengths $>\sim$300 nm the amount of multiple scattering increases because of decreasing ozone absorption, which explains that at this point the scalar radiative transfer error increases. The magnitude and sign of the scalar error depend on the scattering angle and the orientation of the scattering plane for the different scattering processes [*Mischenko et al.*, 1994], which explains the solar zenith angle dependence in Figure 4.

[24] The right panel of Figure 4 shows the ozone profile errors due to the scalar radiative transfer error. The maximum profile error is about 5% in the stratosphere and 70% in the troposphere (solar zenith angle = 25°, albedo = 0.1). The results clearly illustrate the importance of using a vector atmospheric radiative transfer model for ozone profile retrieval from GOME. Obviously, this is also true for ozone profile retrieval from comparable instruments, like SCIAMACHY (nadir mode), GOME-2, and OMI. For limb viewing geometry the scalar error is evaluated by *Oikarinen* [2001].

4. Modeling the Polarization-Sensitive Measurement

4.1. Forward Model Description

[25] A correct and natural way to treat instrument polarization sensitivity is to use the polarization-sensitive measurements I_{pol} as elements of the measurement vector **y** in (1) and take polarization into account in modeling the transport of radiation through both the atmosphere and the instrument. In this way the two polarization dependent errors discussed in section 3 are eliminated.

[26] In general, the forward model in (1) is nonlinear. A linearization by a Taylor expansion,

$$\mathbf{F}(\mathbf{x}) = \mathbf{F}(\mathbf{x}_o) + \frac{\partial \mathbf{F}}{\partial \mathbf{x}}(\mathbf{x}_o)[\mathbf{x} - \mathbf{x}_o] + O(\mathbf{x} - \mathbf{x}_o)^2, \qquad (16)$$

where $O(\mathbf{x} - \mathbf{x}_o)^2$ denotes higher order terms, allows one to solve the inversion problem iteratively with standard fitting methods like the least squares method, the optimal estimation method [*Rodgers*, 1976], and Phillips-Tikhonov regularization [*Phillips*, 1962; *Tikhonov*, 1963]. So, for the direct treatment of the polarization sensitivity of the instrument we need a forward model that simulates $\mathbf{y} = [I_{\text{pol}}(\lambda_1), \ldots, I_{\text{pol}}(\lambda_M)]^T$, and its derivatives with respect to the atmospheric state vector **x**. For this purpose we employ a linearized vector radiative transfer model which uses the Gauss-Seidel iteration technique for solving the vector radiative transfer equation and calculates the derivative of the forward model in (16) by the use of the forward-adjoint radiative perturbation theory [*Marshuk*, 1964; *Box et al.*, 1989]. A detailed description of the model is given by

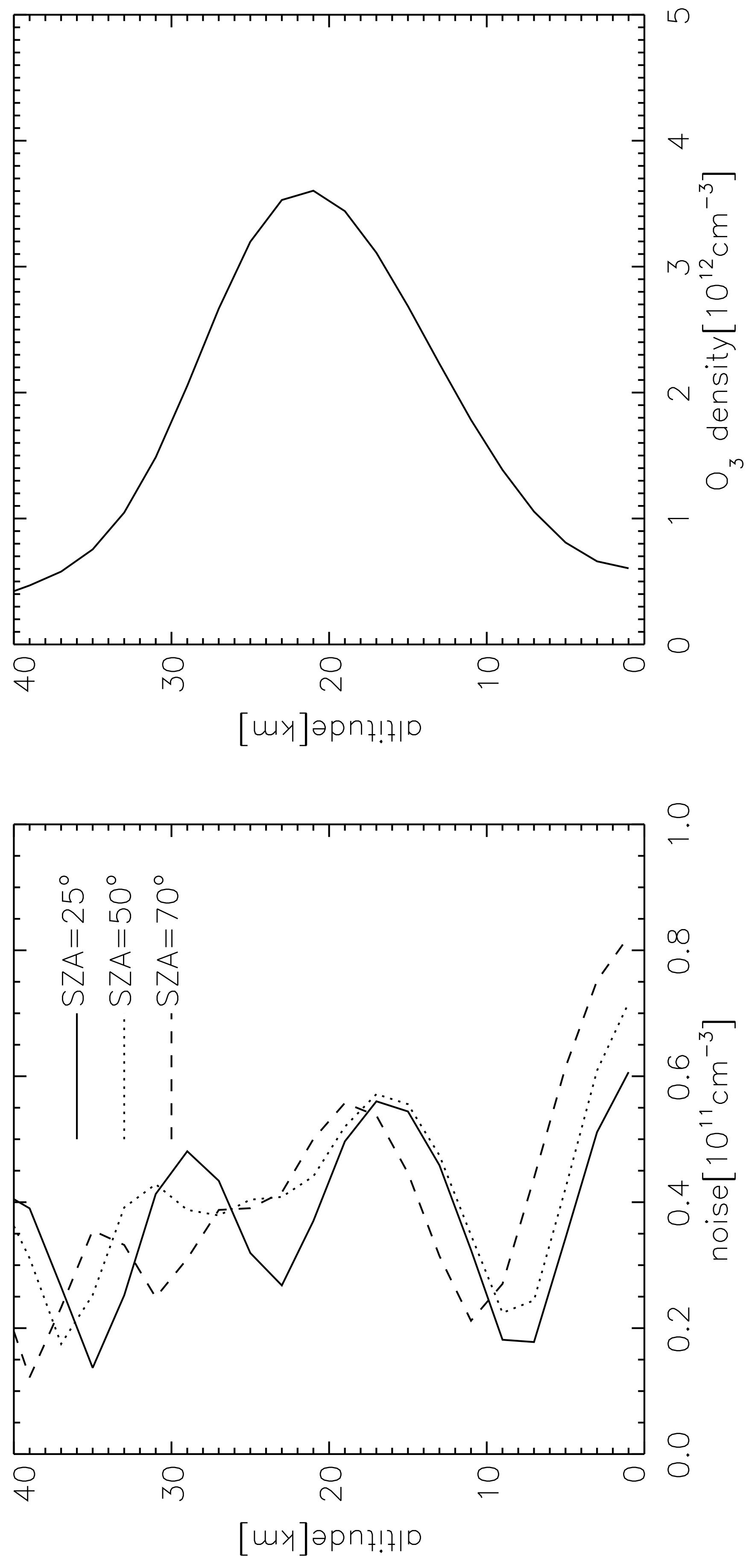

Figure 3. (left) Retrieval noise and (right) profile corresponding to the retrievals of Figures 2 and 4.

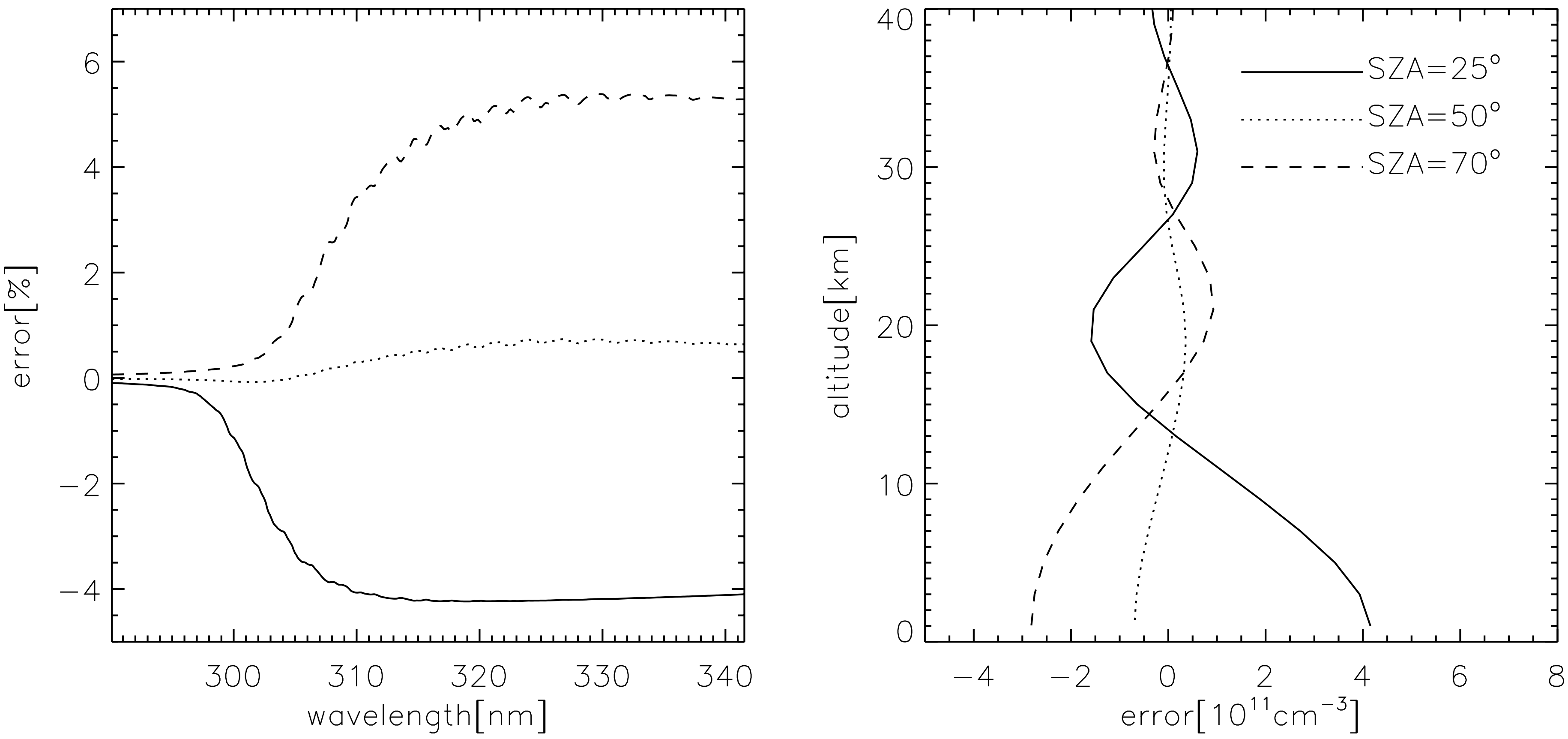

Figure 4. (left) Relative error in modeled radiance if the scalar approximation for radiative transfer is used and (right) the effect of this error on the retrieved ozone profiles. Same model atmosphere, surface albedo, and viewing scenario as in Figure 1. The corresponding ozone profile is depicted in the right panel of Figure 3. For the calculations a Gaussian measurement noise is assumed that is representative for GOME.

Hasekamp and Landgraf [2002a]. Here, we summarize aspects of the model that are of particular importance for the purpose of this paper.

[27] In order to model the polarization-sensitive measurement I_{pol} we need to calculate the intensity vector $\mathbf{I}$ for a given state of the atmosphere, which is found by solving the radiative transfer equation in its forward formulation

$$\hat{\mathbf{L}}\,\mathbf{I} = \mathbf{S}, \tag{17}$$

where the transport operator $\hat{\mathbf{L}}$ is an integro-differential operator. For the form of $\hat{\mathbf{L}}$ see the papers of *Hasekamp and Landgraf* [2002a] and *Landgraf et al.* [2001, 2002]. The radiation source $\mathbf{S}$ is in the ultraviolet and visible part of the spectrum given by the unpolarized sunlight that illuminates the top of the model atmosphere:

$$\mathbf{S} = \mu_o \delta(z - z_{top}) \delta(\Omega - \Omega_o) [F_o, 0, 0, 0]^T. \tag{18}$$

Here F_o is the extraterrestrial flux per unit area perpendicular to the incoming solar beam, δ denotes the Dirac delta function, z is altitude and $\Omega = (\mu, \varphi)$ where μ is the cosine of the solar zenith angle and φ is the azimuthal angle. Furthermore, z_{top} indicates the height of the model atmosphere, and $\Omega_o = (-\mu_o, \varphi_o)$ describes the geometry of the incoming solar beam.

[28] The solution of equation (17) yields the intensity vector field. In this paper we are interested in a certain radiative effect E of this field from which the polarization-sensitive measurement I_{pol} can be simulated. The radiative effect can be extracted from the vector intensity field $\mathbf{I}$ with a suited response vector function $\mathbf{R}$ via

$$E = \langle \mathbf{R}, \mathbf{I} \rangle, \tag{19}$$

where the inner product of two arbitrary vector functions $\mathbf{a}$ and $\mathbf{b}$ is defined by

$$\langle \mathbf{a}, \mathbf{b} \rangle = \int_0^{z_{top}} dz \int_{4\pi} d\Omega\, \mathbf{a}^T \mathbf{b}, \tag{20}$$

with $d\Omega = d\mu\, d\varphi$ and the integration is over full solid angle and altitude range of the model atmosphere. As follows from (7), the suited response vector function for the problem considered here is given by

$$\mathbf{R}(z, \boldsymbol{\Omega}) = \delta(z - z_{top})\ \delta(\boldsymbol{\Omega} - \boldsymbol{\Omega}_v)\ \ [1, m_{12}, m_{13}, m_{14}]^T, \tag{21}$$

where $\boldsymbol{\Omega}_v = (\mu_v, \varphi_v)$ denotes the viewing direction of the instrument. The polarization-sensitive measurement I_{pol} can be simulated by integration of the radiative effect E over all viewing angles that correspond to a ground pixel and by taking the instrument's slit function and the sampling over detector pixel into account.

[29] In order to calculate the derivatives of the forward model with respect to the state vector $\mathbf{x}$ by means of the forward-adjoint perturbation theory [*Marchuk*, 1964; *Box et al.*, 1989], we need besides the solution of the forward radiative transfer problem (17), also the solution of the adjoint problem,

$$\hat{\mathbf{L}}^\dagger \mathbf{I}^\dagger = \mathbf{S}^\dagger. \tag{22}$$

Here, $\mathbf{I}^\dagger$ is the adjoint intensity vector field and $\hat{\mathbf{L}}^\dagger$ is the adjoint operator [*Carter et al.*, 1978; *Hasekamp and Landgraf*, 2002a]. For the adjoint source $\mathbf{S}^\dagger$ in principle any source can be chosen, defining the specific adjoint problem.

[30] If we take the response vector function $\mathbf{R}$ in (21) as the adjoint source, the derivative of the radiative effect E with respect to the kth element x_k of $\mathbf{x}$ can be calculated from:

$$\frac{\partial E}{\partial x_k} = -\frac{1}{\Delta x_k} \langle \mathbf{I}^\dagger, \Delta\hat{\mathbf{L}}\,\mathbf{I} \rangle, \tag{23}$$

where $\Delta\hat{\mathbf{L}}$ is a change in $\hat{\mathbf{L}}$ caused by a change Δx_k in x_k. From (23) it follows that for the needed derivatives of the forward model with respect to any element x_k of the state vector, only two radiative transfer problems have to be solved: the forward problem (17) and the adjoint problem (22) with the response vector function (21) as the adjoint source. Thus, the forward-adjoint perturbation theory for vector radiative transfer allows one to implement in a natural way the polarization sensitivity of the instrument in the forward model. With such a forward model there is no need for using polarization-corrected radiances.

[31] The use of our vector radiative transfer code for ozone profile retrieval takes about a factor 10 more computation time than the corresponding scalar version of our code. For (near) real time data processing, a possible option to speed the calculations up is to only perform vector calculations at a small set of wavelengths and use an interpolation to obtain values for q, the scalar error, and the partial derivatives at intermediate wavelengths. Such an interpolation scheme may make use of the fact that both the relative Stokes parameter q and the scalar radiative transfer error are strongly correlated with the fraction of light that is multiply scattered. The latter can be estimated well by the scalar radiative transfer approach. Another approach to treat the scalar radiative transfer error is to use a lookup table. This approach will be followed for ozone profile retrieval from OMI.

4.2. Validation With Ozonesondes

[32] The proposed approach for dealing with instrument polarization sensitivity is applied to ozone profile retrieval from GOME. For the inversion we use the model described by *Hasekamp and Landgraf* [2001]. A summary of the inversion method is given in Appendix A of this paper. Retrieved profiles for the period April 1996 till June 1998 are validated with 123 coincident ozonesondes, launched at Payerne, Switzerland (lat. 46.8, lon. 7.0).

[33] Before comparing the retrieved ozone profiles with high resolution profiles that are measured by ozonesondes, we degrade the ozonesonde profiles to the same vertical resolution as the GOME profiles. This is done with the corresponding averaging kernel (see Appendix A of this paper). In this way we eliminate differences between sonde- and GOME profiles that result from limitations of the measurement concept of GOME, and thus can focus on differences caused by errors in the measurement and forward model, respectively. For an example of the averaging kernel for the used inversion method, see *Hasekamp and Landgraf* [2001].

[34] In Figures 5–7 the retrieved ozone densities at 21, 15, and 7 km are compared in a time series with

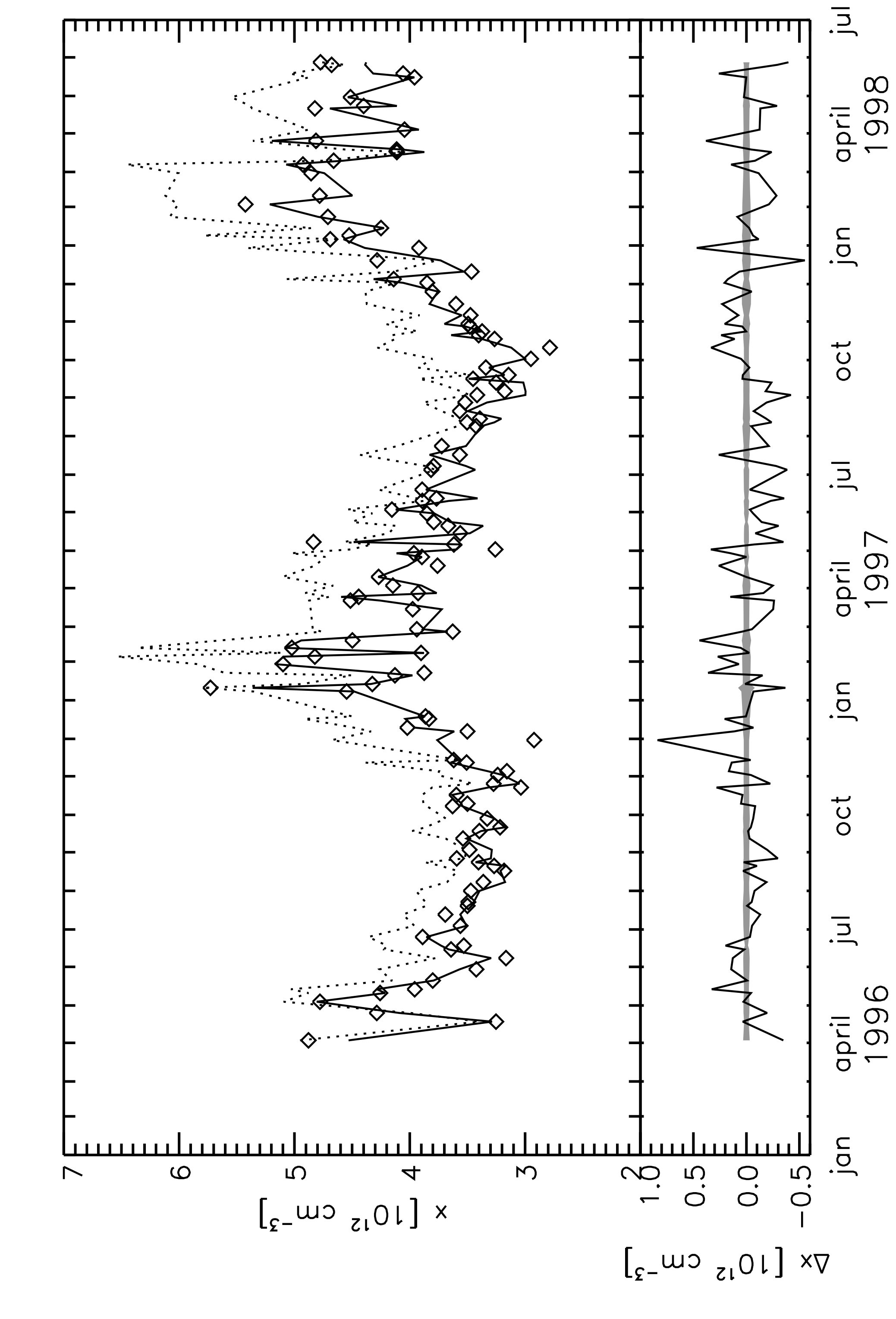

Figure 5. (upper panel) Time series comparison of GOME profiles with ozonesonde profiles at 21 km altitude for Payerne, Switzerland. The dotted line represents the sonde profile on the retrieval grid and the solid line shows the sonde profile multiplied with the averaging kernel. For validation the solid line has to compared with the diamonds which indicate the retrieved ozone values for collocated ground pixels. In total 123 collocations are shown. (lower panel) Difference between the concentration of the smoothed sonde and the retrieval. The gray area shows the 1σ error band of the retrieval noise.

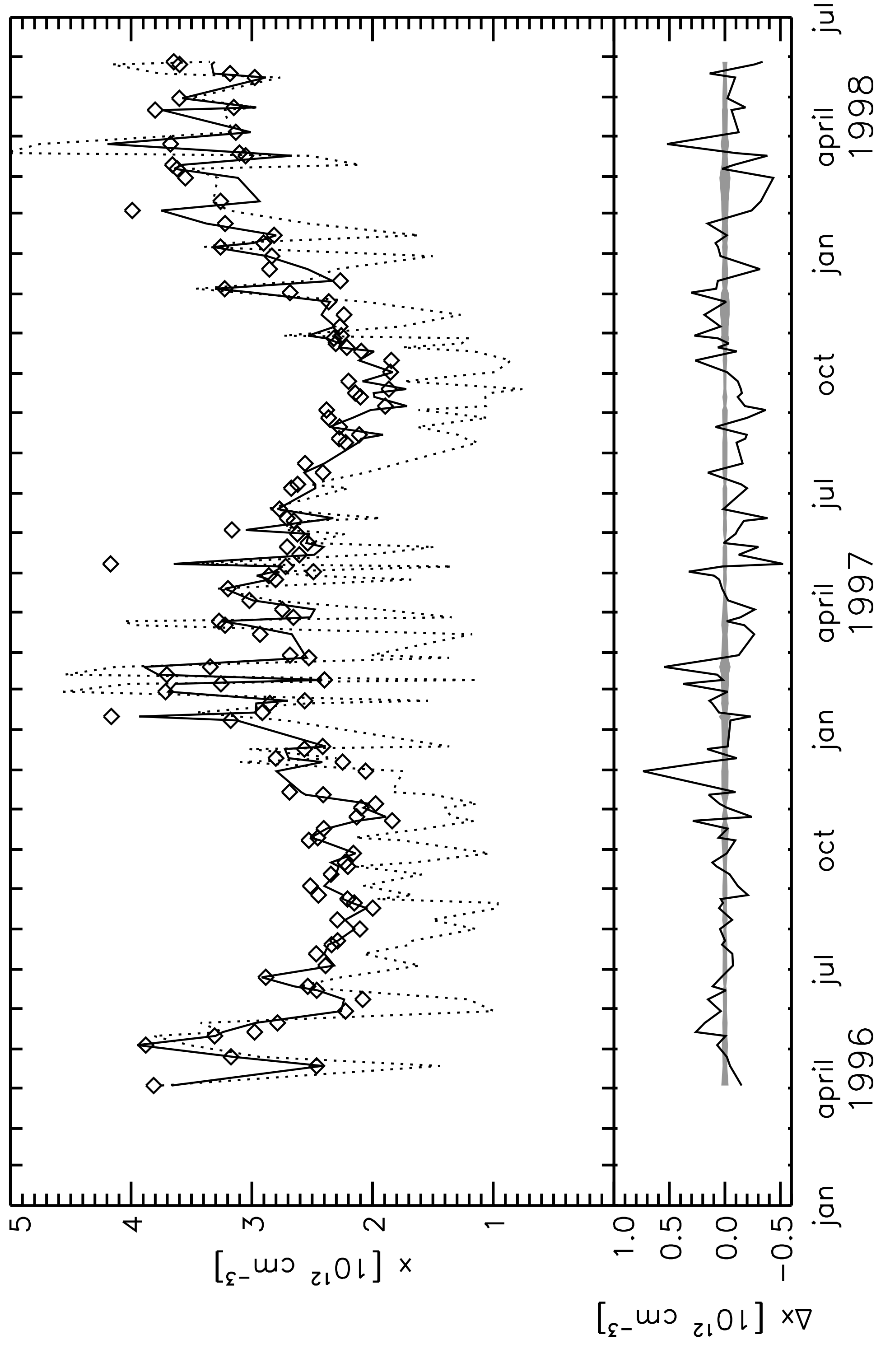

Figure 6. Same as Figure 5 but at 15 km altitude.

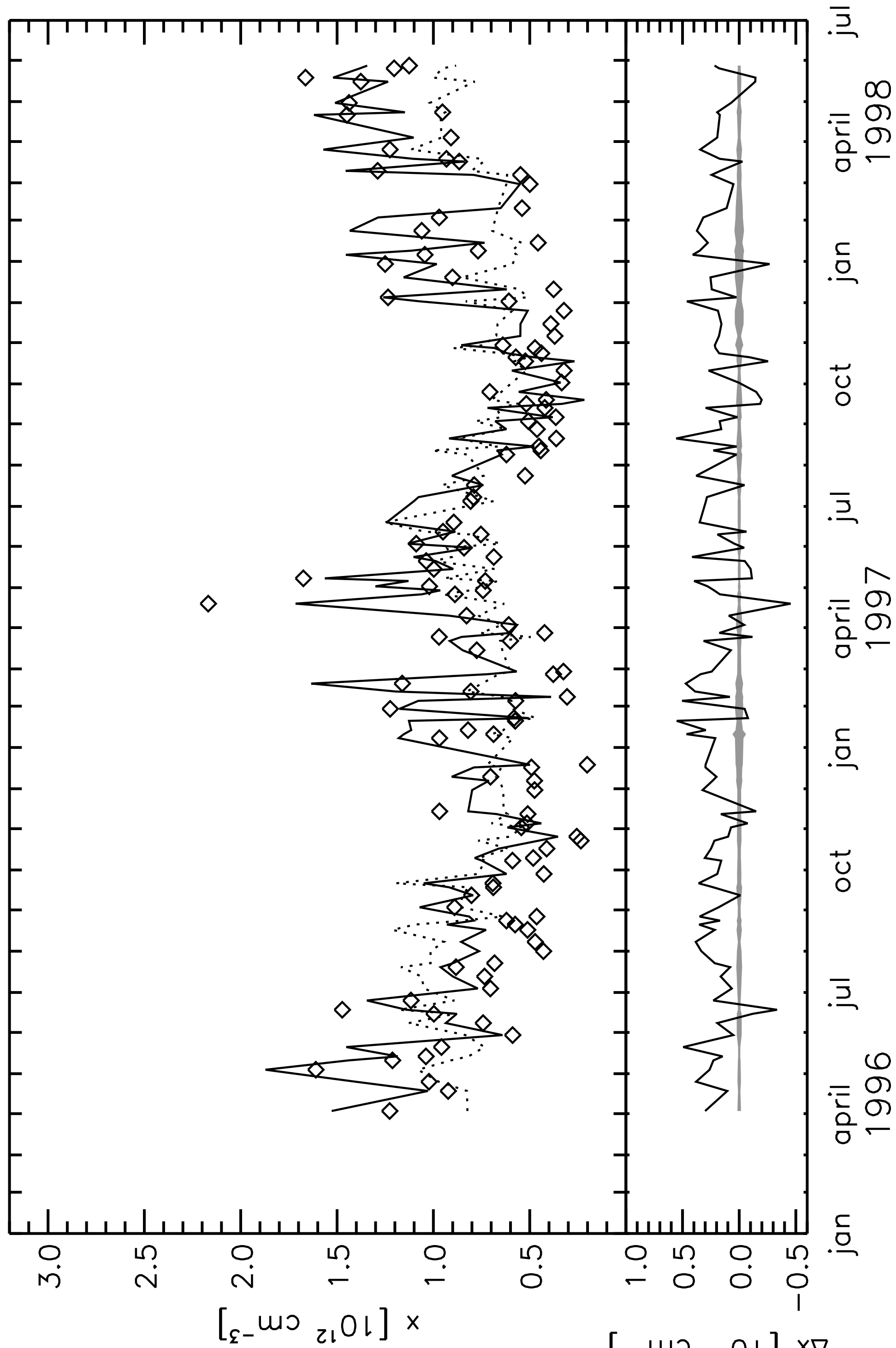

Figure 7. Same as Figure 5 but at 7 km altitude.

corresponding smoothed ozonesonde measurements. The comparison shows good agreement and no clear seasonal deviation of the retrieved profiles can be identified. However, the differences are significant larger than the retrieval noise, which is also shown in the time series. This may partly be caused by ozone variations within the GOME ground pixel, but probably the differences are for a large part due to, for example, radiometric calibration errors [*van der A*, 2001], and the treatment of clouds as an enhanced surface reflection in the retrieval. Additionally, Figures 5–7 show the densities of the nonsmoothed ozonesonde measurements. These data illustrate that the daily and seasonal ozone variability that is detected by the high resolution ozonesonde measurements can also be clearly seen in the smoothed sonde profiles, although the smoothing (A2) damps these features.

[35] Currently, the standard approach to ozone profile retrieval from GOME is to use polarization corrected radiances and a scalar radiative transfer model [*Munro et al.*, 1998; *Hoogen et al.*, 1999; *Hasekamp and Landgraf*, 2001]. In order to compare the here proposed approach with this standard approach, Figure 8 shows the mean retrieved profile of the time series and the root-mean-square (RMS) difference between the GOME ozone profiles and the ozonesonde profiles, for the two approaches. The RMS retrieval noise is also plotted in Figure 8. The improvement in RMS difference due to the proper treatment of polarization in the forward model is about a factor 1.5 in the stratosphere as well as in the troposphere. This improvement is obviously significant, as it is a factor 5–10 larger than the retrieval noise. Additionally, Figure 8 shows the RMS difference for retrievals performed on polarization-corrected radiances, but with use of a vector model for atmospheric radiative transfer. The differences in Figure 8 clearly demonstrate that using the vector approach for radiative transfer through the atmosphere only is not enough, which means that the extension to polarization modeling through the instrument is essential.

[36] *Hasekamp and Landgraf* [2001] presented a validation of ozone profiles retrieved from GOME using polarization-corrected radiances and a scalar model for atmospheric radiative transfer. A clear correlation was found between the GOME-sonde differences and solar zenith angle. The results of section 3 of this paper suggest that this correlation may be caused by noncorrect treatment of polarization (see Figures 2 and 4). For the GOME-sonde comparisons of this paper the corresponding solar zenith angle dependence of the differences at an altitude of 7 km is shown in Figure 9 for the standard approach, and in Figure 10 for the approach presented in this paper. From these figures it follows that the inclusion of polarization in the model for radiation transport through the atmosphere and the instrument, reduces significantly the solar zenith angle dependent error. This is an essential improvement, because a solar zenith angle dependent error may severely complicate the interpretation of the retrieved profiles. For a satellite instrument like GOME that flies in a sun-synchronous orbit, this type of error can easily be misinterpreted as a seasonal ozone variation.

[37] In summary, the results of the validation shown in this section indicate that polarization related errors cause a significant problem in current GOME ozone profile retrieval schemes. These problems can be solved by the use of a forward model that includes polarization.

5. Conclusion

[38] In this paper we have demonstrated that polarization sensitivity of satellite instruments can be handled in a proper way within ozone profile retrieval algorithms, by using a forward model that includes polarization in the modeling of radiation transport both through the atmosphere and the instrument. The approach is also applicable to the retrieval of other atmospheric parameters. So, in general there is no need to correct measurements for instrument polarization sensitivity. The vector approach for atmospheric radiative transfer, which is needed in order to model the radiance correctly, allows a straightforward extension to modeling polarized radiation transport through the instrument.

[39] The standard approach in atmospheric constituent retrieval algorithms for GOME, SCIAMACHY, and GOME-2, is to use polarization-corrected radiances and a scalar model for atmospheric radiative transfer. The polarization correction is based on broadband polarization measurements and thus contains errors in parts of the spectrum where the state of polarization is changing rapidly with wavelength. Furthermore, the scalar approximation of radiative transfer introduces significant errors in the forward model. For ozone profile retrieval from GOME measurements we studied the two polarization dependent errors in measurement and forward model respectively, and calculated the effect of these errors on the retrieved ozone profile. The error due to the polarization correction can be as large as 12% in the stratosphere and 200% in the troposphere, while the error due to the use of a scalar atmospheric radiative transfer model can be 5% in the stratosphere and 70% in the troposphere. Both errors can be eliminated with the use of a vector forward model.

[40] A validation of ozone profiles retrieved from GOME with coincident ozonesonde profiles showed a significant improvement due to the here proposed method for polarization treatment. Compared to the standard approach mentioned above, the total improvement in root-mean-square difference is about a factor 1.5 in the stratosphere as well as in the troposphere. Not only the root-mean-square difference between GOME ozone profiles and sonde profiles is reduced, also a solar zenith angle dependence in the differences is reduced significantly.

[41] The approach presented in this paper is aimed at the interpretation of measurements of GOME, SCIAMACHY, and GOME-2. The proper treatment of the polarization sensitivity of these instruments overcomes an apparent disadvantage with regard to OMI, which scrambles the polarization of the observed light, yielding an effective polarization insensitive instrument. In fact, the polarization measurements of GOME, SCIAMACHY, and especially GOME-2 are not needed for the purpose of polarization correction but should be used to extract additional atmospheric information on, for example, aerosols [*Mishchenko*

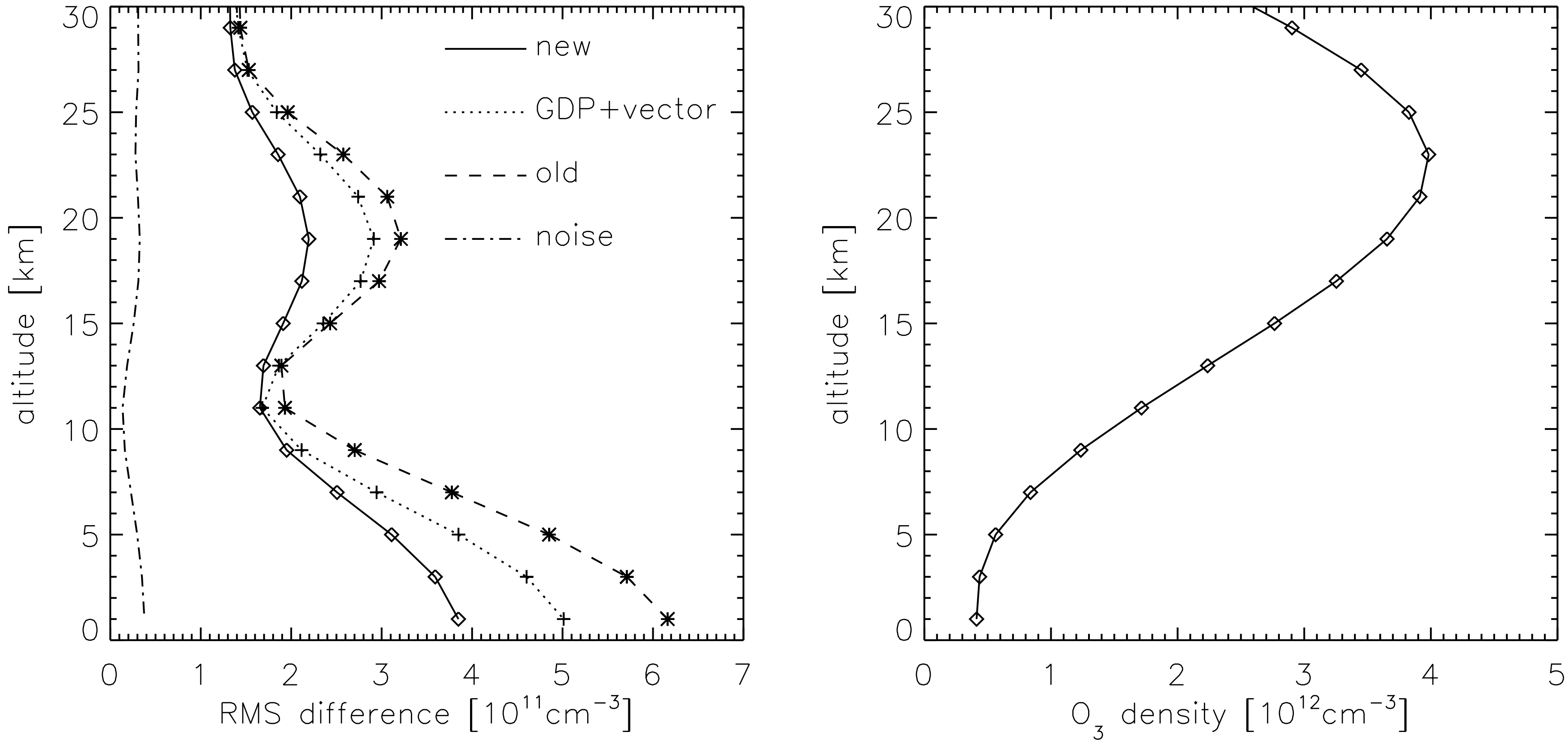

Figure 8. (left) Root-mean-square difference between ozone profiles retrieved from GOME and ozonesonde profiles. The shown retrieval noise (dot-dashed line) is representative for all approaches. (right) Mean retrieved profile. "new" corresponds to the proposed approach. "old" corresponds to the approach using polarization corrected radiances and a scalar radiative transfer model, and "GDP + vector" corresponds to the approach using polarization corrected radiances but a vector radiative transfer model. For the comparison the ozonesonde profiles have been smoothed by the averaging kernel.

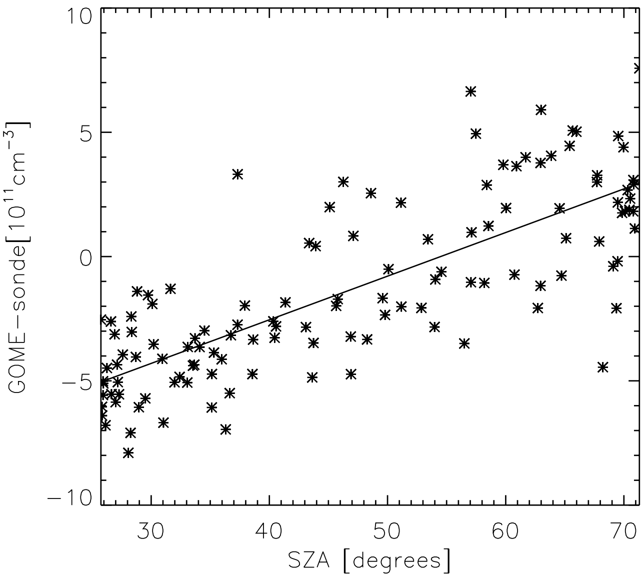

Figure 9. Solar zenith angle (SZA) dependence of the difference between GOME- and ozonesonde profiles at 7 km (see also Figure 7), for a retrieval procedure that makes use of the GDP polarization corrected radiances and a scalar radiative transfer model. The solid line corresponds to the linear regression. For the comparison the ozonesonde profiles have been smoothed by the corresponding averaging kernel.

and Travis, 1997] and tropospheric ozone [*Hasekamp and Landgraf*, 2002b].

Appendix A: Description of Ozone Profile Retrieval Method

[42] The retrieval of ozone profiles from satellite measurements of backscattered sunlight represents an ill-posed problem, because the measured spectrum is insensitive to fine scale altitude structures in the ozone profile. In this paper we treat the ill-posed problem of ozone profile retrieval using the Phillips-Tikhonov regularization technique [*Phillips*, 1962; *Tikhonov*, 1963] including minimization of the first derivative norm of the profile in addition to the least squares condition, viz.

$$\mathbf{x} = \min_{\mathbf{x}} \left(\|\mathbf{S}_y^{-\frac{1}{2}}(\mathbf{F}(\mathbf{x}) - \mathbf{y})\|^2 + \gamma^2 \ \|\mathbf{H}\mathbf{x}\|^2 \right), \tag{A1}$$

where $\mathbf{S}_y$ is the noise covariance matrix of $\mathbf{y}$, $\mathbf{H}$ is the discrete representation of the first derivative and γ is a regularization parameter balancing the minimization of the least squares condition and the minimization of the first derivative norm. The rationale behind the minimization of the first derivative norm as a side constraint is that the measurement $\mathbf{y}$ is insensitive to fine scale structures of the ozone profile. These vertical structures do not influence the residual norm but strongly influence the first derivative norm. The regularization parameter γ should be chosen such that the retrieved profile contains all vertical structures that influence the measurement while the structures to which the measurement is insensitive should be filtered out. Such a value of the regularization parameter is found from the L curve [*Hansen and O'Leary*, 1993]. The L curve is a parametric plot of $\|\mathbf{H}\mathbf{x}\|\ (\gamma)$ versus $\|\mathbf{S}_y^{-\frac{1}{2}}(\mathbf{F}(\mathbf{x}) - \mathbf{y})\|(\gamma)$, which has an L-shaped corner. The corner of the L curve corresponds to the optimum value of the regularization parameter, because at this point a decrease of γ would not improve the residual norm but would lead to a strong increase of the first derivative norm, while on the other hand, an increase in γ would make the residual norm larger. Note that for the retrieval procedure not the absolute values of the elements of $\mathbf{S}_y$ are important but only the relative spectral behavior. In this paper we assume uncorrelated Gaussian noise.

[43] Because the measured spectrum is insensitive to fine scale vertical ozone structures, the retrieved profile $\mathbf{x}_{\rm ret}$ is a smoothed version of the true profile $\mathbf{x}_{\rm true}$,

$$\mathbf{x}_{\rm ret} = \mathbf{A}\ \mathbf{x}_{\rm true} + \mathbf{e}_x, \tag{A2}$$

where $\mathbf{e}_x$ is the profile error caused by errors in the forward model and measurement, and $\mathbf{A}$ is the averaging kernel [*Rodgers*, 2000]. For the application and validation of the retrieval result, (A2) should always be kept in mind. The retrieved profile should not be considered as a true profile but as a smoothed profile that depends on the true profile as in (A2). Thus, for the validation of a retrieved profile $\mathbf{x}_{\rm ret}$ with an ozonesonde profile $\mathbf{x}_{\rm son}$ we compare $\mathbf{x}_{\rm ret}$ with the smoothed sonde profile $\mathbf{A}\ \mathbf{x}_{\rm son}$.

[44] An important feature of the Phillips-Tikhonov regularization method is that it does not depend on climatological information about the ozone profile (i.e., it does not need an a priori profile and corresponding covariance matrix). In the context of ozone profile retrieval from GOME data the method is discussed in detail by *Hasekamp and Landgraf* [2001]. There also a comparison is made with the commonly used optimal estimation method *Rodgers* [1976]. Other atmospheric applications of Phillips-

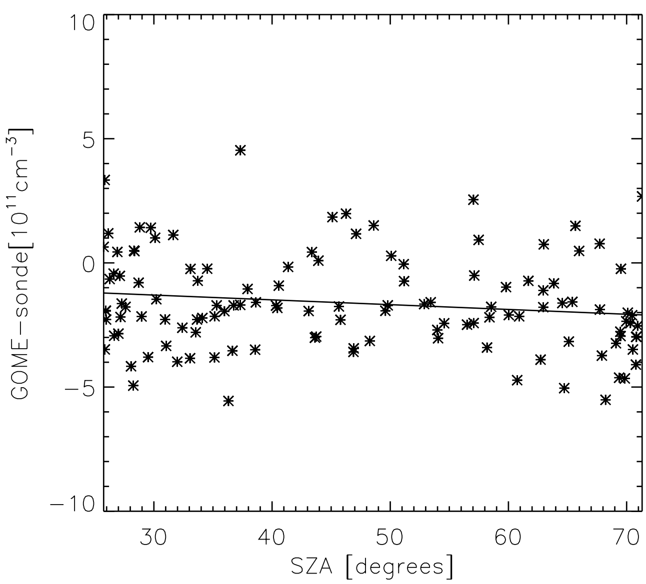

Figure 10. Same as in Figure 9 but for a retrieval procedure where the polarization-sensitive measurement is modeled.

Tikhonov regularization in combination with the L curve have been reported by, for example, *Schimpf and Schreier* [1997] and *Liu et al.* [1999].

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