

# Fast NO<sub>2</sub> retrievals from Odin-OSIRIS limb scatter measurements

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**Abstract.** The feasibility of retrieving vertical profiles of NO<sub>2</sub> from space-based measurements of limb scattered sunlight has been demonstrated using several different data sets since the 1980's. The NO<sub>2</sub> data product routinely retrieved from measurements made by the Optical Spectrograph and InfraRed Imaging System (OSIRIS) instrument onboard the Odin satellite uses a spectral fitting technique over the 437 to 451 nm range, over which there are 36 individual wavelength measurements. In this work we present a proof of concept technique for the retrieval of NO<sub>2</sub> using only 4 of the 36 OSIRIS measurements in this wavelength range, which reduces the computational cost by almost an order of magnitude. The method is an adaptation of a triplet analysis technique that is currently used for the OSIRIS retrievals of ozone at Chappuis band wavelengths. The results obtained are shown to be in very good agreement with the spectral fit method, and provide an important alternative for applications where the computational burden is very high. Additionally this provides a baseline for future instrument design in terms of cost effectiveness and reducing spectral range requirements.

## 1 Introduction

Observations of stratospheric NO<sub>2</sub> began in the 1970's from ground-based zenith-sky measurements (Brewer et al., 1973) and solar occultation from high altitude balloon (Kerr and McElroy, 1976). The pioneering measurement of the stratospheric NO<sub>2</sub> profile from limb scattering geometry actually occurred from space, with the Solar Mesosphere Explorer (Mount et al., 1984). Depending on the orbit, radiances were measured at two wavelengths of equal NO<sub>2</sub> cross-section,

428.7 and 431.8 nm, or two wavelengths with differing NO<sub>2</sub> cross-section, 439.3 and 442.3 nm (Naudet et al., 1987). McElroy (1988) used measurements from balloon limb scattering geometry at five wavelengths in the 437 to 450 nm range to better account for interfering ozone absorption in the retrieval of NO<sub>2</sub>. Recently, measurement of the NO<sub>2</sub> vertical profile by balloon-borne limb scattering was revived by Weidner et al. (2005) who used a spectrometer and retrieved NO<sub>2</sub> by spectral fitting in the 400–450 nm range. Space-based solar occultation measurements of NO<sub>2</sub> were first made by the Stratospheric Aerosol and Gas Experiment (SAGE) II instrument in 1984 (Cunnold et al., 1991).

With the launch of OSIRIS (Llewellyn et al., 2004) on the Odin satellite in 2001 (Murtagh et al., 2002), the information available for the retrieval of NO<sub>2</sub> from space-based limb scatter measurements improved drastically in terms of spectral resolution and range. Sioris et al. (2003) presented the first NO<sub>2</sub> profiles from OSIRIS. Furthermore, Haley et al. (2004) developed a two-step retrieval algorithm for NO<sub>2</sub> relying on differential optical absorption spectroscopy and the maximum a posteriori estimator (see also Haley and Brohede, 2007). Profiles from this algorithm were validated (Brohede et al., 2007b) and used to develop a climatology of stratospheric NO<sub>2</sub> in terms of mean and standard deviation, as a function of latitude, altitude, local solar time and month (Brohede et al., 2007a). The capability of OSIRIS to measure NO<sub>2</sub> in the upper troposphere was best demonstrated by Sioris et al. (2007b), who found that enhancements in NO<sub>2</sub> in this region are predominantly from lightning-generated NO<sub>x</sub>. Additionally Sioris et al. (2007a) illustrated Rossby wave breaking with OSIRIS NO<sub>2</sub> near the edge of the Antarctic polar vortex. Recently, Tukiainen et al. (2008) completed the development of the Finnish modified onion-peeling algorithm which determines the ozone profile in a first onion peeling stage and the NO<sub>2</sub> profile in the second iteration.



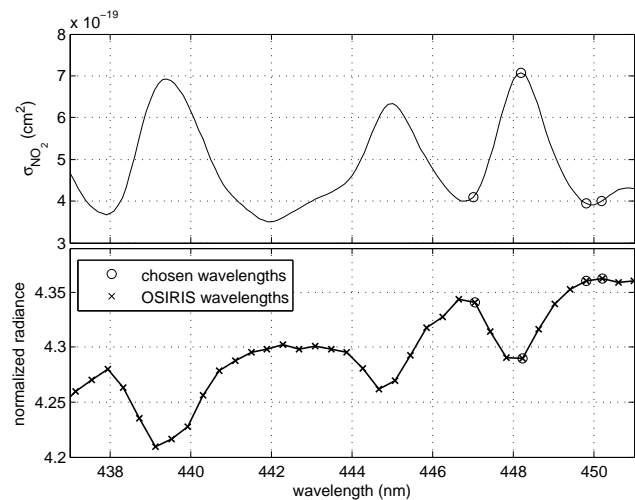
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NO<sub>2</sub> has been retrieved from SCIAMACHY limb scattering measurements operationally by the Germany Aerospace Center (Doicu et al., 2007a,b) and by scientific research groups at universities of Bremen (Eichmann et al., 2004; Rozanov et al., 2005; von Savigny et al., 2005), Harvard (Sioris et al., 2004), and also at the Max Planck Institute for Chemistry (Kühl et al., 2008; Puķite et al., 2010). The measurements of the scientific research groups have been independently validated by Butz et al. (2006). All of the SCIAMACHY retrievals use a spectral fitting technique over a window of 30 nm or wider in the 420–495 nm range.

The large spectral range measured by current instruments such as OSIRIS and SCIAMACHY, obtained using CCDs or photodiode array detectors, generally leads to greater sensitivity, better precision, and an improved ability to remove interfering signals. Nonetheless, in this work an alternative OSIRIS NO<sub>2</sub> algorithm is presented that uses only a small subset of these wavelengths. An algorithm that can achieve results close to that from the full spectrum is an attractive alternative for several reasons. Instruments based on the diffraction grating and solid-state detector design are expensive, complicated and require a large downlink bandwidth. Their throughput is also limited since optical entrance slits must be narrow to achieve the desired resolution. The addition of a few discrete channels in the NO<sub>2</sub> absorption region is an alternative for instruments designed for high-resolution in other spectral regions, for example, an O<sub>3</sub>-BrO-OCIO instrument. Alternatively it can provide an efficient correction for interference due to NO<sub>2</sub> in an instrument dedicated to measurements of another species, such as the Ozone Mapping and Profiling Suite (OMPS) scheduled for launch on the NPP satellite (Flynn et al., 2009). This is also an important alternative to dramatically reduce the computational time associated with a retrieval application where the computational burden is very high as can be the case with a two dimensional inversion.

## 2 The methodology

The retrieval technique we have employed uses a measurement vector that is constructed based on the technique used for space-based limb scatter retrievals of stratospheric ozone in the Chappuis band as discussed in a number of publications including Flittner et al. (2000), von Savigny et al. (2003), (Roth et al., 2007) and (Degenstein et al., 2009). It is also similar to that used for NO<sub>2</sub> retrievals from the ground-based Brewer MkIV instrument (Cede et al., 2006). In these retrieval schemes, the measurement vector is a combination of vertical limb scatter radiance profiles at a small number of wavelengths. In this work, we generalize the previously published “triplet” technique, discussed in the ozone retrieval algorithms referenced above, to use the radiance at 4 strategically chosen wavelengths measured by OSIRIS to construct the measurement vector elements. A Multiplicative



**Fig. 1.** The NO<sub>2</sub> cross section and the measured OSIRIS spectrum in the wavelength range from 437 nm to 451 nm. The 4 wavelengths used in this technique are indicated with circle markers at the long wavelength end. The lower panel shows an OSIRIS 23 km tangent altitude spectrum in the same wavelength range after it has been normalized by the OSIRIS spectrum measured along a 45 km tangent altitude line of sight. OSIRIS pixels are indicated with x markers.

Algebraic Reconstruction Technique, which has been successfully used for the retrieval of other species from the OSIRIS measurements (Bourassa et al., 2007; Degenstein et al., 2009), is used to iteratively solve for the NO<sub>2</sub> profile. The SASKTRAN model (Bourassa et al., 2008), which is a spherical successive orders radiative transfer code, is used for the forward model in this work.

Haley and Brohede (2007) provide a detailed sensitivity analysis for the OSIRIS NO<sub>2</sub> retrieval and the results presented there regarding systematic error from forward model parameter error and pointing uncertainty apply to this retrieval technique. The NO<sub>2</sub> absorption cross sections are taken from tabulated values reported by Burrows et al. (1998) with a linearly interpolated temperature dependence and convolved to the OSIRIS slit function.

### 2.1 The measurement vector

The construction of the measurement vector relies on the fact that the structure in the NO<sub>2</sub> cross section is relatively broad with respect to the 1 nm OSIRIS spectral resolution. A typical NO<sub>2</sub> cross section is shown in the upper panel of Fig. 1 in the spectral window from 437 to 451 nm. This spectral range is used in the official OSIRIS version 3.0 NO<sub>2</sub> data product, which is based on the spectral fitting technique presented in Haley et al. (2004) and Haley and Brohede (2007). The lower panel of Fig. 1 shows the spectrum measured by OSIRIS along a line of sight tangent at 25 km altitude. For illustrative purposes, this spectrum has been normalized in a wavelength-by-wavelength sense with a spectrum measured

along a line of sight tangent at 45 km altitude. Each of the 36 OSIRIS pixels in this window are marked by x's on the spectrum, all of which are used in the spectral fitting technique for the official version 3.0 data product. In this work, we have strategically chosen 4 OSIRIS pixels as a minimum set of wavelengths to use for an NO<sub>2</sub> retrieval. These are indicated by the circles in each plot.

The familiar form of the typical triplet measurement vector, as commonly used in the retrievals of ozone from limb scattered sunlight noted above, is defined as

$$y_j = \ln \left( \frac{\sqrt{I_j(\lambda_1) I_j(\lambda_2)}}{I_j(\lambda_3)} \right). \quad (1)$$

In this equation the measurement vector element  $y_j$ , that corresponds to a tangent altitude denoted by the subscript  $j$ , is calculated using three radiance measurements, two of which are reference wavelengths that are typically weakly absorbing ( $\lambda_1$  and  $\lambda_2$ ), and a third at a strongly absorbing wavelength ( $\lambda_3$ ). Equation (1) can be written equivalently as

$$y_j = 0.5 \ln (I_j(\lambda_1)) + 0.5 \ln (I_j(\lambda_2)) - 1.0 \ln (I_j(\lambda_3)). \quad (2)$$

This form of the triplet measurement vector is also common in the literature and is relevant to this work. In this form, there is a coefficient that multiplies the natural log of each radiance measurement, i.e. 0.5, 0.5 and  $-1.0$ . Positive coefficients scale the reference or weakly absorbing wavelengths and negative coefficients scale the absorbing wavelength. Also, the sum of all coefficients is identically zero, as the reference wavelength coefficients sum to 1.0 and the absorbing wavelength coefficients sum to  $-1.0$ .

Following this logic, Eq. (2) can be generalized for  $n$  radiance measurements and written as,

$$y_j = p_1 \ln (I_j(\lambda_1)) + p_2 \ln (I_j(\lambda_2)) + \dots \quad (3)$$

$$+ q_1 \ln (I_j(\lambda_m)) + q_2 \ln (I_j(\lambda_{m+1})) + \dots$$

where wavelengths 1 to  $m-1$  are the reference wavelengths and the coefficients

$$\sum_i p_i = 1 \quad (4)$$

and wavelengths  $m$  to  $n$  are the absorbing wavelengths and

$$\sum_i q_i = -1 \quad (5)$$

such that the sum over all coefficients is zero.

The measurement vector used in this work is constructed using four radiance measurements. Three of these are weakly absorbing reference wavelengths:  $\lambda_1 = 447.04$  nm,  $\lambda_2 = 449.81$  nm and  $\lambda_3 = 450.21$  nm and are located at local minimums in the cross section as shown in Fig. 1. A single absorbing wavelength,  $\lambda_4 = 448.23$  nm, is located at the local

maximum of the cross section. The measurement vector is constructed as

$$y_j = 0.5 \ln (I_j(\lambda_1)) + 0.25 \ln (I_j(\lambda_2)) \quad (6)$$

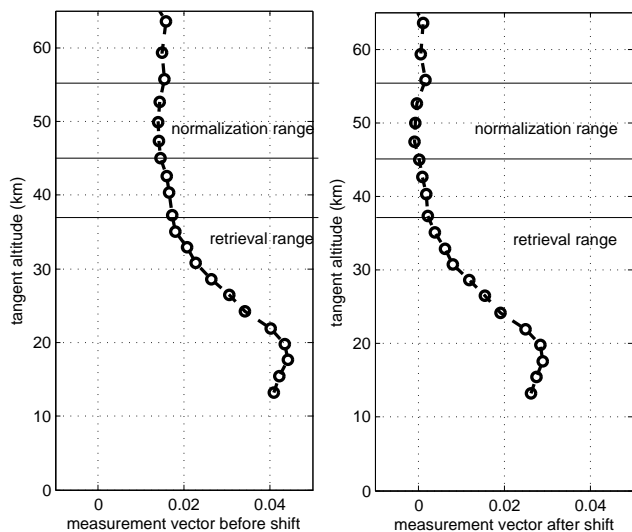
$$+ 0.25 \ln (I_j(\lambda_3)) - 1.0 \ln (I_j(\lambda_4)).$$

This effectively states that the measurement is the difference of the weighted average of the log of the radiance at the three reference wavelengths and the log of the wavelength at the local peak in the NO<sub>2</sub> cross section. These wavelengths are strategically chosen so as to maximize the sensitivity to NO<sub>2</sub> with a very small subset of radiance measurements. As with the triplet technique, the reference wavelengths are chosen on either side of the absorption feature. With the absorbing wavelength at the peak, the triplet effectively is a measure of the depth of the absorption. We have chosen to add a fourth wavelength so as to increase the number of measurements and systematically reduce the effect of random noise. However, using wavelengths between the local maximum and minimum values of the absorption feature, i.e. within the feature, serves only to reduce the sensitivity of the measurement vector. For example, if multiple wavelengths around the peak were incorporated into the measurement vector, the effective depth of the absorption feature is decreased through the averaging. Thus because the minimum in the cross section is relatively wide on the long wavelength side of the absorption feature, we can add an additional reference wavelength without decreasing the effective depth. These two reference wavelengths on the long wavelength side of the absorption peak are averaged with equal weight with respect to the reference on the short side wavelength of the peak.

or a specific application, it would be possible to study an optimal spectral resolution for this retrieval technique. However, it is clear that as a first order estimate the slit function must be at least a few times narrower than the absorption feature being used in order to implement this technique.

### The normalization technique

Typically, measurement vector elements for limb scatter retrievals are normalized by a measurement at a higher tangent altitude. This removes the requirement for an absolute calibration and decreases systematic effects due to uncertainty in forward model parameters such as the effective reflectance or albedo term (see for example von Savigny et al., 2003). For this work we have not used a direct division by higher tangent altitude measurements. As shown in Eq. (6) the radiance at each tangent height is used directly. For a typical OSIRIS scan, the result of applying Eq. (6) is shown in the left hand panel of Fig. 2. In this case, the measurement vector approaches a constant non-zero value at high tangent altitude. Fluctuations about this value are due to measurement noise. By definition, a normalized measurement vector is identically zero at the normalization altitude as each term simplifies to the logarithm of 1.0. Due to this logarithmic



**Fig. 2.** The measurement vectors. The vector shown in the left hand panel is calculated using Eq. (6). The vector shown in the right hand panel is shifted by a constant such that the average value is zero over the normalization range.

nature of the measurement vector, the normalization can be viewed as an offset term, constant with tangent altitude, that drives the measurement vector to zero at the normalization altitude.

Since the radiances at high tangent altitude can contain significant noise due to decreasing limb signal, the noise in a single measurement used for normalization can systematically affect the entire profile as it results in an offset of the measurement vector. For this reason, rather than normalize with a single measurement, we calculate the average value of the measurement vector over a range of high tangent altitudes and then offset the entire measurement vector, in a negative sense, by that average value. The overall effect is similar to a high altitude normalization and the measurement vector approaches zero at high tangent altitude; however, the impact of noise is reduced by using multiple measurements. This is shown in the right panel of Fig. 2, where the normalization range includes all measurements used to construct the average value used for the offset.

This same procedure is applied to the measured and the modelled vectors during the retrieval process. It has almost no impact on computational efficiency of the algorithm as the vast majority of computation time is spent forward modelling the radiance profiles for different wavelengths; additional lines of sight at a given wavelength require almost no additional computational time as the most intensive portion is the calculation of the diffuse, or multiple scattering component of the radiation field, which is coupled between all altitudes. Thus the time required to obtain the solution for an additional line of sight at a higher or lower tangent altitude from the same satellite position is negligible.

## 2.2 The MART inversion

The Multiplicative Algebraic Reconstruction Technique (MART) equation, used to iteratively update the state parameter based on the forward modelled measurement vector,

$$\mathbf{x}_i^{(n+1)} = \mathbf{x}_i^{(n)} \sum_j \left( \frac{y_j^{\text{measured}}}{y_j^{\text{modelled}}} W_{ji} \right), \quad (7)$$

has been described in application to limb scatter in many previous publications (see for example Roth et al., 2007, Bourassa et al., 2007, Degenstein et al., 2009). It has also been used to retrieve 1.27  $\mu\text{m}$  Oxygen InfraRed Atmospheric Band emissions in the mesosphere (Degenstein et al., 2003). The application of this equation is straight forward. The ratio of the measured and modelled measurement vector  $y_j$ , where  $j$  indicates tangent altitude, it is used to retrieve a the state parameter vector  $x_i$ , where the  $i$  indicates altitude. Of course in this case,  $x_i$  is the vertical profile of NO<sub>2</sub>.

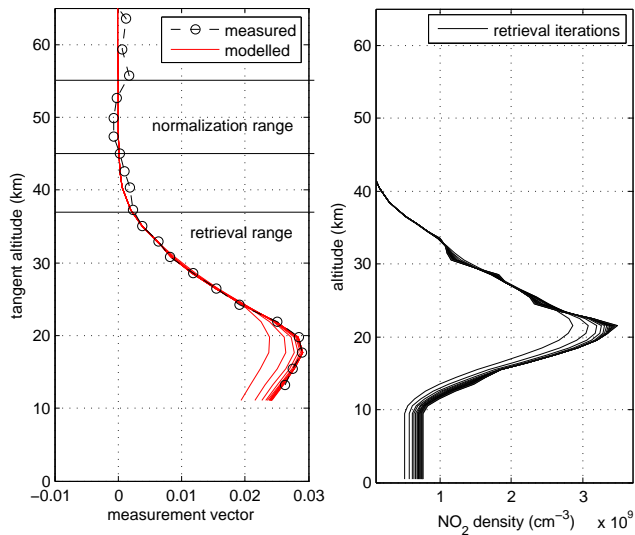
In this work, three measurement vector elements are considered important to the retrieval at any given altitude,  $i$ . These are the measurement vector elements corresponding to tangent altitudes through the spherical shell at altitude  $i$  and the next two lower tangent altitude vector elements. These two measurements have significant path lengths through the shell at altitude  $i$  on the near and far sides of the tangent point. The weights  $W_{ij}$  associated with these three vector elements are 0.5, 0.3 and 0.2 where the weight decreases with decreasing tangent altitude. For a full discussion of this see Bourassa et al. (2007).

A photochemical box model is used to generate the initial guess profile. This is the same profile that is used as the a priori profile for the optimal estimation retrieval used for the spectral fitting in the official version 3.0 product (Brohede et al., 2007a). The measurement vector for a typical scan and the last 10 of 15 iterations of the retrieval are shown in Fig. 3.

## 3 Results

The results from the retrieval using the methodology outlined above can be directly compared with the official OSIRIS version 3.0 NO<sub>2</sub> data product. As a proof of concept for the purposes of this work a typical full day of OSIRIS measurements is analyzed. This is the illuminated portion of 16 full orbits of measurements, each including approximately 60 vertical scans. On 22 April 2002, the sunlit portion of the Odin orbit covers almost the entire Northern Hemisphere. The ascending node of the orbit is at local dusk; as the satellite passes over the northernmost part of the orbit, the time is local noon. The descending node is at local dawn. As NO<sub>2</sub> is photochemically active, the comparisons between the two methods must be done such that local time is synchronized for all retrievals going into a single average.

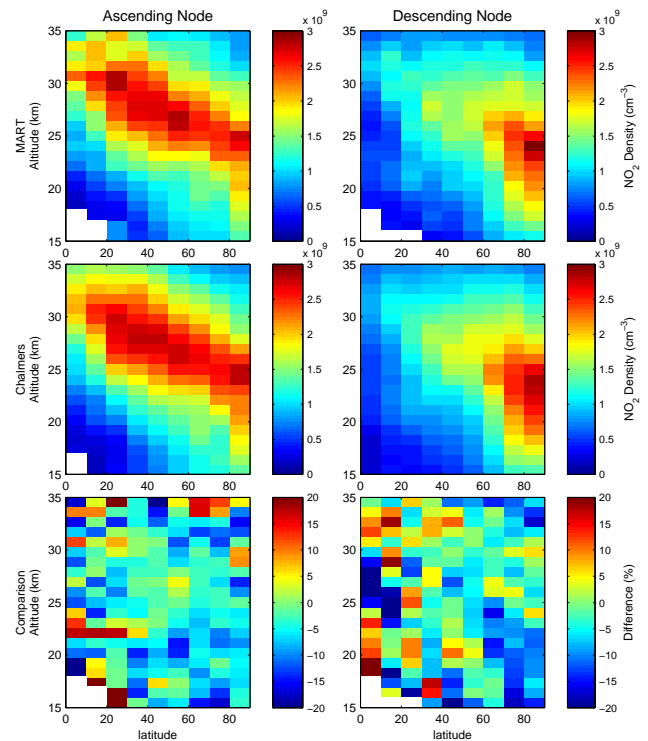
On a typical desktop processing machine the retrieval for a single scan takes approximately 1 min to perform. The



**Fig. 3.** The convergence of the measurement vector and the retrieved NO<sub>2</sub> profile. The left panel shows the measured vector and its modelled value for the last 10 of 15 iterations of the retrieval. Also indicated in this panel are the range over which the vector is normalized and the range over which the NO<sub>2</sub> is retrieved.

SASKTRAN model is multi-threaded and thus has the ability to use multiple-core machines efficiently (Bourassa et al., 2008). However, this factor aside, the vast majority of CPU time required for the retrieval is taken by the SASKTRAN forward model calculations of the radiances. This is typical of most retrieval schemes for limb scatter measurements due to the large number of calculations required to account for the multiple scattering. While it is true that the addition of more measurements at individual wavelengths will improve the retrieval precision, in the interest of processing time, using only 4 wavelengths, as opposed to 36 for the official version 3.0 product, is approximately 9 times faster.

Figure 4 shows the comparison between the official OSIRIS version 3.0 NO<sub>2</sub> retrievals, which use the entire spectral window, and those calculated using the methodology outlined above using the radiance at the 4 indicated wavelengths. The upper panels show our results for measurements collected on the descending track or morning twilight node of the Odin orbit on the left and for measurements collected on the ascending track or morning twilight node of the Odin orbit on the right. All retrievals for the 16 orbits on 22 April 2002, are binned in 10° of latitude. The northernmost bin contains only data from 80 to 82° as the inclination of the Odin orbit is 98°. Recall that the Odin orbit is such that the low latitude bins represent measurements made at local dusk or dawn and the most northern bins represent data collected near local noon. A rapid sweep between local times occurs around 70° latitude. The middle panel of Fig. 4 shows the same but for the OSIRIS version 3.0 retrieval. The comparison is qualitatively very good and on

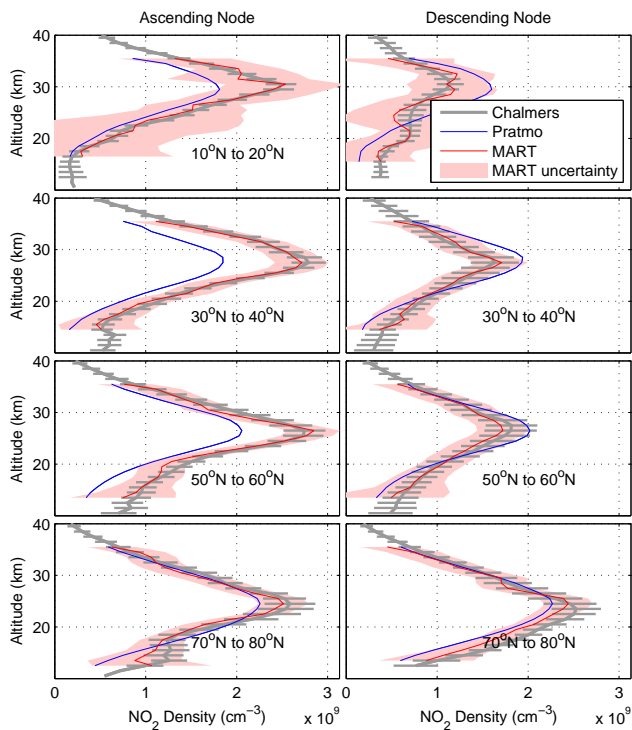


**Fig. 4.** A daily average of our retrievals (top) compared with the official OSIRIS version 3.0 retrievals (middle). The left column shows measurements made during the descending track of the Odin orbit, i.e. measurements at local morning twilight. The right column shows measurements from the ascending track of the orbit during local evening twilight. The bottom panels show the percent difference between the two retrieved data sets.

average there are very few differences between the values as retrieved from identical radiance measurements but with these different techniques.

The lower panel of Fig. 4 shows the percent difference between the results shown in the upper two panels. The percent difference is determined as the difference between our results minus the official version divided by the official version and expressed as a percentage. Only at the altitude extremes is there any significant difference between the results obtained from the two methods. The upper two panels of this figure show how the peak altitude of the NO<sub>2</sub> profile decreases in altitude as latitude increases or as the local time progresses. Generally, the percent difference between the two methods is less than 10% near the peak of the NO<sub>2</sub> number density profile. The differences between the two retrievals increases in a random sense at low latitudes and high altitudes. This is reflected in the precision estimate of the retrieval, which is discussed further below.

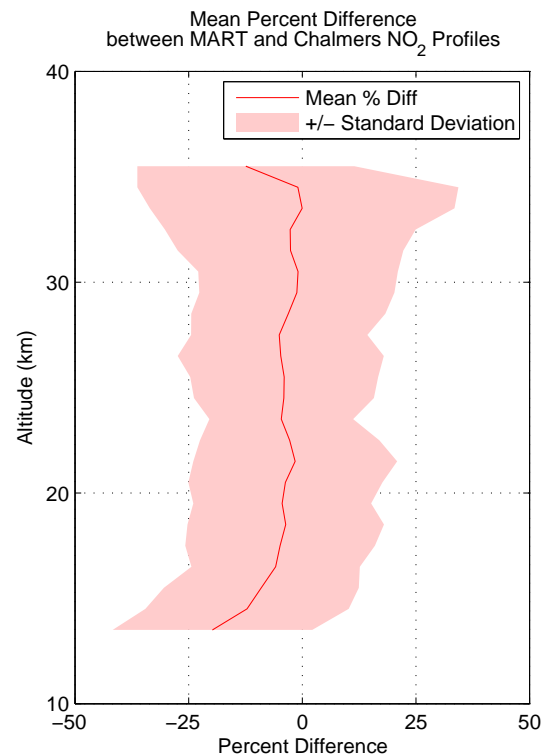
As a further illustration of the quality of the data retrieved using this technique Fig. 5 shows the mean vertical profiles taken from the panels of Fig. 4. Also included on this figure are the initial guess for the MART technique, which as



**Fig. 5.** Selected zonal mean profiles from one day of OSIRIS measurements. Each panel compares the zonal average of our retrievals and the official OSIRIS version 3.0 product. Included in each panel is the zonal average of the apriori/initial guess profile (labeled Pratio). In both cases, the uncertainty represents the average uncertainty for a profile in the latitude bin. The range of solar zenith angles, in degrees, in each bin, from low to high latitude are: 81–87, 75–80, 70–74, and 67–69.

mentioned previously is the apriori for the version 3.0 retrievals. For all latitudes and local time conditions there is good agreement between the results retrieved using the two different methods. This figure also shows a numerical estimate of the precision of the MART retrieval (in the red shading) and the error estimate from the official NO<sub>2</sub> product (in gray bars). The error bars shown are the average uncertainty for each of the average profiles and not the error in the average profile. Thus these error bars show the typical error for a single retrieved profile in each of the latitude bins. As expected, the uncertainty in the MART retrievals is systematically larger than that in the official product. However, for mid and high latitudes where the signal to noise is high due to the fact that the solar zenith angles is smaller, i.e. less than 80° at the tangent point, the MART error bars are much less than a factor of two larger those of the official product. However, in the tropics where the signal to noise is decreased, the MART uncertainty can be several times larger than the uncertainty in the official product.

As a final comparison between the two retrieval methods, Fig. 6 shows the statistics for the comparison of all individual profiles retrieved during this same day of measurements. The



**Fig. 6.** The mean of the percent difference and associated standard deviation between MART and version 3.0 retrievals for all profiles measured on the same day of OSIRIS measurements.

comparison is relatively good; the mean of the percent difference from each comparison shows a tendency toward a small negative bias that is less than 5% at altitudes between 20 and 30 km, and increases with decreasing altitude below 20 km. The standard deviation of the comparison is approximately 20% at all altitudes, which reflects the size of the error bar in the retrieval as shown in Fig. 5.

#### 4 Conclusions

The retrievals of NO<sub>2</sub> produced by the MART technique outlined here, using radiances measured at 4 wavelengths, compare very well with those obtained by the official OSIRIS version 3.0 product, which uses 36 wavelengths in a spectral fitting inversion. The precision of the retrieval is comparable to the version 3.0 product for high latitudes where the signal to noise ratio is high. At lower latitudes the uncertainty of the MART retrievals is somewhat larger and increases the differences between the two sets of results. The statistics for the comparison of a full day of OSIRIS measurements shows essentially no mean bias and a standard deviation that is approximately 20%.

The computation time required for our retrievals is reduced by almost an order of magnitude from a spectral window fitting technique. This provides a viable alternative

method for tomographic inversion or as a fast and self consistent correction for NO<sub>2</sub> as an interfering species in a measurement and retrieval focused on another species.

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