

Stratospheric and tropospheric NO₂ observed by SCIAMACHY: first results

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Abstract

Observations from the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) onboard Envisat are used to quantify the vertical distribution of stratospheric nitrogen dioxide (11–41 km) from limb scattering geometry and the tropospheric column from limb–nadir matching. SCIAMACHY is validated with spatially and temporally coincident observations from Optical Spectrograph and Infrared Imager System (OSIRIS) and from Halogen Occultation Experiment (HALOE). Comparison with the Canadian Middle Atmosphere Model provides a further check of consistency. Errors in pointing are detected and corrected using the recently developed ‘spectral knee’ technique. An instrumental artifact, presumably a tangent-height-dependent wavelength drift, is causing fine spectral structure but can be taken into account as a pseudo-absorber in the least squares fitting. Extending the fitting window to longer wavelengths than those currently employed by other optical satellite-borne NO₂ sensors allows for the retrieval to penetrate the lower stratosphere. This extended fitting window is used to retrieve lower stratospheric NO₂ in the denoxified Antarctic polar vortex. Finally, tropospheric NO₂ columns, retrieved from limb–nadir matching, are presented.

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1. Introduction

Remote sensing of stratospheric NO₂ profiles from limb scattered radiance spectra has developed into an accurate technique (Sioris et al., *J. Geophys. Res.*, 2003; S2003 hereafter) that provides global coverage in the sunlit hemisphere. Significant improvements have been achieved over early efforts from balloon (McElroy, 1988) and space (Mount et al., 1984).

The Optical Spectrograph and Infrared Imager System (OSIRIS, Llewellyn et al., 1997) onboard the Odin satellite began measuring stratospheric profiles of NO₂ from the limb scatter technique in the summer of 2001. Early validation results (S2003) have been quite encouraging.

Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) (Bovens-

mann et al., 1999) is onboard the near-polar-orbiting sun-synchronous EnviSat-1 platform, launched March 1, 2002. Its descending node is at 10:00 local time (LT). SCIAMACHY alternates between nadir and limb viewing in such a way that a limb swath and subsequent nadir swath match spatially and are separated in time by 7 min. A SCIAMACHY limb scan typically consists of up to eight azimuth steps for each of 34 elevation steps. Nine hundred and sixty kilometres are sampled in the azimuthal direction during each elevation step (Bovensmann et al., 1999). The vertical sampling is 3.3 km starting at a tangent height (TH) of –3 km and extending up to ~100 km. Spectrally, SCIAMACHY is an 8-channel grating spectrometer covering the 240–2380 nm range. Channel 3 (394–620 nm) covers the portion of the spectrum where NO₂ exhibits the largest differential absorption structure. The calibration features of the instrument and the expected signal to noise ratio (SNR) of the limb and nadir measurements are presented in Bovensmann et al. (1999). The radiometric accuracy and precision are 2–4% and <1% (Bovensmann et al., 1999)

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and the wavelength calibration is typically accurate to <9 e-4 nm. First limb radiance profiles and spectra are illustrated in Kaiser et al. (2004).

Several previous retrievals of tropospheric NO₂ (e.g. Martin et al., 2002) assumed a zonally invariant stratospheric NO₂ column to produce a tropospheric residual. Limb–nadir matching has the potential to improve significantly on the separation of stratospheric and tropospheric NO₂, especially during winter where the zonal invariance assumption is not valid due to dynamical disturbances. Tropospheric NO₂ columns can be used to improve our understanding of tropospheric ozone.

In this paper, we compare coincident NO₂ profile retrievals from these two state-of-the-art limb scatter instruments during the summer of 2002. A comparison of SCIAMACHY with Halogen Occultation Experiment (HALOE) is also shown. We present sample meridional cross sections of stratospheric NO₂ processed from selected orbits of SCIAMACHY data and tropospheric residual columns from limb–nadir matching.

2. Method

The retrieval algorithm builds on previous work (S2003). Only differences from that method will be described.

3. Data analysis

Limb radiance spectra are calibrated using Envi-View2.0.6 to remove the stray light contribution, dark current, etalon effects, and memory effect. The pixel-to-pixel gain and wavelength calibrations are also applied.

Unless noted otherwise, the NO₂ fitting window for the observations is 434.7–449.8 nm, similar to the OSIRIS window (S2003). The fitting window was extended to 494.9 nm to improve the penetration of the lower atmosphere by incorporating more optically thin wavelengths and to increase the window-integrated SNR. The disadvantage of such a large fitting window is that the radiative transfer model (RTM) simulations during the inversion must also cover a large wavelength range unless some subset of the fitting window can be found which gives equivalent slant column density (SCD) profiles.

SCIAMACHY limb radiance spectra measured at different azimuths of the same elevation step are co-added. This is necessary to achieve a good SNR especially at high THs. Spectra from THs between 10 and 43 km are normalized with a co-added high altitude reference (I_0) consisting of all tangent heights between 43 and 70 km.

A Ring effect correction is omitted for both SCIAMACHY and OSIRIS data analysis in this work. Normalization with the high altitude reference removes the Ring signature to a very good approximation (S2003). This is apparent from the lack of spectral fitting residuals at the wavelengths of Fraunhofer line cores.

Fraunhofer structure also fails to cancel out perfectly with the high TH reference because of the ‘tilt’ pseudo-absorber (S2003). A ‘tilt’ differential spectrum (S2003) has been simulated for channel 3 of SCIAMACHY assuming the spectral point spread function is a Gaussian with full-width at half-maximum (FWHM) of 0.44 nm (Bovensmann et al., 1999). The simulation uses the Chance–Kurucz solar spectrum from the solar database of MODTRAN4 (Berk et al., 1999; Chance and Spurr, 1997; Kurucz, 1995 and references therein). Including ‘tilt’ reduces the uncertainty of the NO₂ SCD by 20–25% (relatively) at a TH of ~ 11 km and solar zenith angle (SZA) of 53° and increases the SCD by $\sim 5\%$ due to a slight anti-correlation (-0.092 ± 0.004) between ‘tilt’ and NO₂, which depends on the temperature of the absorbing NO₂. The importance of ‘tilt’ increases monotonically with the optical thickness along the line of sight.

The inclusion of another pseudo-absorber has clearly improved the fit quality. The pseudo-absorber is a differential version of the ratio of the observed spectrum at the lowest TH in the Fraunhofer reference (i.e., ~ 46 km) divided by the co-added high altitude reference (46–70 km). This pseudo-absorber, which is calculated every limb scan, has substantial fine differential structure and appears to fit with less uncertainty at tangent heights closer to the lowest reference altitude (TH = 46 km). The radiance spectrum at TH = 46 km has a negligible, and more importantly, similar NO₂ signature to that of the co-added I_0 , thus this pseudo-absorber does not exhibit an NO₂ signature. The spectral structure is evidently not of atmospheric origin. The high-altitude Fraunhofer reference spectrum and the spectra at the tangent heights of interest are assumed to have no wavelength shift between them in the least squares fitting routine. However, Savigny (2002) found evidence of a TH-dependent wavelength shift in channel 2 and 3. Thus, it is likely that this pseudo-absorber is due to small wavelength shifts that are related to changes in the way the detector arrays are illuminated by the optics over the course of a limb scan. NO₂ SCD uncertainties are reduced by up to a factor of 2 with the inclusion of this pseudo-absorber.

The absorbers included in the fitting process are NO₂ and O₂ at 203, 223, and 243 K (Bogumil et al., 2003), which have cross-section uncertainties of 3.4% and 3.1%, respectively (Orphal, 2003), and the O₂–O₂ collision complex (Greenblatt et al., 1990). The O₂–O₂ collision complex becomes important only at THs ≤ 30 km when the extended fitting window is used. A switch from

a quadratic (S2003) to a cubic as a closure polynomial has now been justified based on the adjusted r^2 of the fit.

3.1. Forward model and inversion algorithm

A versatile, pseudo-spherical limb RTM (McLinden et al., 2002) is used as a forward model. A “successive orders of scattering” technique is used to solve the radiative transfer equation. This model is much faster than MODTRAN4 for aerosol-free cases and more accurate in that the variation of the SZA along the tangent path is taken into account. It also contains a more extensive database of model atmospheres (McLinden et al., 2002). The database has a latitudinal resolution of 10° and monthly temporal resolution. The RTM has been modified to read in updated guesses of the NO₂ number density profile produced by the inversion algorithm. It is run in aerosol-free mode since the retrieval is quite insensitive to the current abundance of stratospheric aerosols (S2003). The model is run in unpolarized mode since the retrieval is not sensitive to the scalar approximation as discussed below. A surface albedo of $A = 0.16$ is assumed for all retrievals, which should lessen the impact of incorrect a priori A as compared to S2003 where $A = 0.04$ was assumed, which is at the low extreme. The higher A value was also chosen to compensate for globally averaged cloud fraction and reflectivity with a single parameter. The radiance simulations include the first five scattering orders.

The forward model has been coupled with a least-squares spectral fitting algorithm to simulate NO₂ SCDs. A cubic is used for closure in the simulations as well. The inversion process consists of iteratively updating the atmospheric profile of NO₂ number density within the retrieval range until the simulated SCDs match the SCDs observed by SCIAMACHY. The observed SCDs are interpolated onto a 4 km grid starting typically at TH = 10 km and going up to TH = 42 km. The number densities are retrieved in a 2-km layered atmosphere.

The inversion approach is based on Chahine’s (1970) relaxation method with modifications to handle nonlinearities introduced by optically thick Rayleigh scattering (S2003). All layers are retrieved simultaneously in this version of the algorithm. In S2003, an iterative onion-peel (IOP) approach was used. The new technique is slightly less stable, particularly for sharply peaked profiles and profiles with multiple laminae but has never generated a profile with significantly negative number densities and always converges. In general, the simultaneous inversion converges more rapidly towards the true profile, particularly at small SZAs, where upwelling radiation becomes significant. For large SZAs, the IOP method is competitive. Since SCIAMACHY observes the atmosphere with small SZAs as a result of its 10 a.m. descending node time, the simultaneous inversion is

clearly preferable. For OSIRIS (S2003), because SZA > 57° , this was not the case.

The first guessed NO₂ number density profile comes from the previous limb scan or in the absence of this, it comes from the MODTRAN4 database of latitudinally and seasonally dependent profiles (Berk et al., 1999). Because the TH range used for the Fraunhofer reference is different for SCIAMACHY than for OSIRIS due to the TH-independent integration time of SCIAMACHY, TH = 54 km was selected as the reference TH in the simulation. This represents the radiance-weighted average TH in the range co-added to obtain I_0 for the normalization of the observations.

The convergence criteria of the inversion have been tightened as compared to S2003. Convergence between modelled and measured SCDs must exist not only at each tangent height, but the sum of the modelled SCDs inversely-weighted by their SCD uncertainty must be within $\pm 1.0\%$ of the measured SCDs inversely weighted by their relative uncertainties. This additional criterion is empirically derived and has been included to prevent small, systematic biases in the retrieval.

3.2. Error analysis

A thorough analysis of errors and sensitivities was performed in S2003. The 1- σ number density precisions are calculated by propagating the standard errors of the NO₂ SCDs through the inversion algorithm (S2003). In this section, we highlight additional tests that have been performed. The impact of using two orders of scattering or less in the forward model can be easily determined using the McLinden et al. (2002) code. Using only two scattering orders appears to give the correct profile to <1% when compared with a profile retrieved with five scattering orders. One order of scattering is clearly insufficient in the lower atmosphere as previously noted (Mount et al., 1984; S2003). The underestimation due to the single scattering approximation reaches 18% at 19 km (for SZA = 58°). The impact of the scalar approximation has also been quantified. It is <1% between 13–39 km. Thus it appears that the full Stokes vector does not need to be included in the forward RTM.

3.3. Tropospheric NO₂ vertical columns

Tropospheric NO₂ vertical column densities (VCDs) are calculated by subtracting the stratospheric vertical column density obtained from limb viewing from the total VCD from nadir geometry obtained with software developed for GOME (Martin et al., 2002) and adapted to SCIAMACHY. The VCD above the altitude range of the limb retrieval, i.e., $z > 42$ km, is assumed to be negligible relative to the underlying tropospheric column. The tropospheric NO₂ VCD presented here also includes a lower stratospheric component ($< 1.05 \times 10^{14}$

mol/cm²) since the limb retrieval is only extended down to the altitude where the NO₂ precision >100%. The extended spectral fitting window has been used to penetrate the lowermost stratosphere. When the extended fitting window is used, the simulation covers the 435–493.8 nm window in 0.52 nm steps. The lower altitude limit of the retrieval range is always ≥ 11 km in the data analyzed here. The stratospheric column from limb scatter is linearly interpolated to the latitude (lat) of the centre of the nadir ground pixel. This accounts for the latitudinal variation of the stratospheric column within a limb swath (but neglects the longitudinal variation). The tropospheric VCD precision is the quadrature sum of the total VCD error and the stratospheric VCD precision from limb.

4. Results

Three OSIRIS-SCIAMACHY coincidences have been selected for a detailed comparison of the NO₂ profiles based on the coincidence criteria used in the early validation of OSIRIS NO₂ (S2003). Comparisons of SCD and number density profiles were made before and after TH correction. The TH correction improves the agreement in all cases. In one case, on August 3, 2002, the agreement of SCD profiles is not adequate even after TH correction. We presume that, even though the coincidence criteria have been met at the top of the retrieval range, some spatially localized gradient in the NO₂ field exists. Also note that the across-track swath of SCIAMACHY and OSIRIS is 960 and 40 km, respectively. Fig. 1 shows one coincidence. In all, $\sim 90\%$ of coincident SCIAMACHY and OSIRIS data points agree in the three coincidences, slightly less than would be expected from 1- σ precision on the correlative mea-

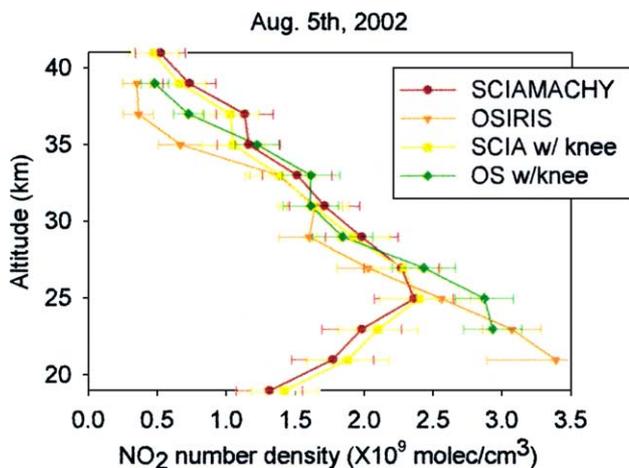


Fig. 1. Coincidence with OSIRIS exists in the 30–40 km range to within 2.5° (lat), 3° in longitude (lon), and 14 min. ‘w/ knee’ indicates that TH correction has been applied.

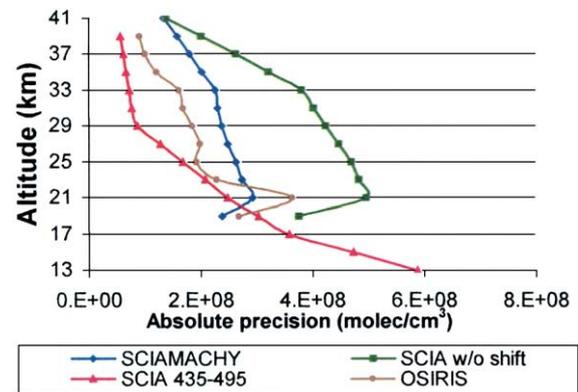


Fig. 2. Retrieval precision averaged over three coincidences, for SCIAMACHY with and without (‘w/o’) TH-dependent wavelength shift correction and for OSIRIS. Also shown is the precision when the extended spectral fitting was used (see text).

surements, and the profile and column agreement are typically 18% and 9%, respectively. Again, we attribute some of the bias to the coincidence criteria being insufficiently stringent for the case on August 3rd.

In Fig. 2, the absolute precision of the two instruments is shown versus altitude. The finer vertical sampling of OSIRIS (~ 2 km) leads to slightly better precision than SCIAMACHY on a common 2 km vertical grid (assuming the same spectral fitting window) even though its SCD uncertainties are generally slightly larger due to its smaller throughput. Also shown in Fig. 2 is the precision using the 435–495 nm fitting window for a profile recorded on October 1. A factor of ~ 2 improvement in the precision is achieved with the extended fitting window, especially at higher tangent heights where the limb radiance begins to decrease exponentially (TH = 30–42 km). The relative precision is as low as 5.5%. At 13 km, the precision is 5.9×10^8 mol/cm³ or 95%.

HALOE continues to be one of the best satellite instruments for validation of NO₂ profiles from limb scatter because HALOE relies on a different technique: IR solar occultation. This provides a very high SNR (Gordley et al., 1996) and the capability to probe the lowermost stratosphere. A sample coincidence is shown in Fig. 3. The SZA coincidence criterion (S2003) was neglected, so a photochemical correction was applied. In a first step, the SCIAMACHY THs are shifted down by the 1.7 km offset determined with the ‘spectral knee’ technique at ~ 305 nm (S2003). The profile is then forecasted to SZA = 90° to match HALOE using a photochemical model (McLinden et al., 2001). After these two additional steps, the columns agree to 7% over the 17–41 km range and the profiles agree at the 8% level, typically.

A comparison with the Canadian Middle Atmosphere Model (CMAM) (de Grandpré et al., 1997) demonstrates the consistency of the SCIAMACHY

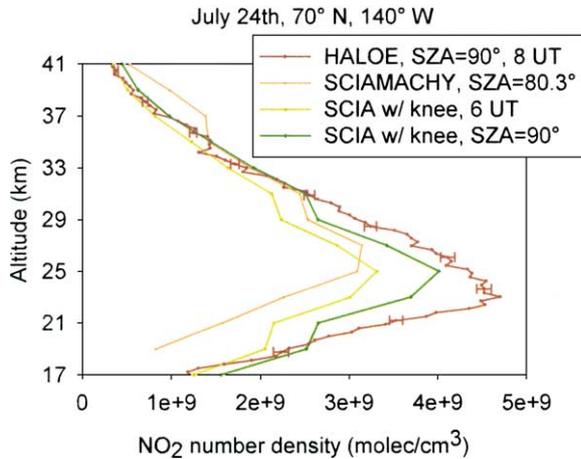


Fig. 3. Coincidence with HALOE at local sunset to 0.3° (lat), 12° (lon), and 124 min. The profile was observed by SCIAMACHY at 10 pm LT. A photochemical model (McLinden et al., 2001) is used to forecast the profile to local sunset to match HALOE.

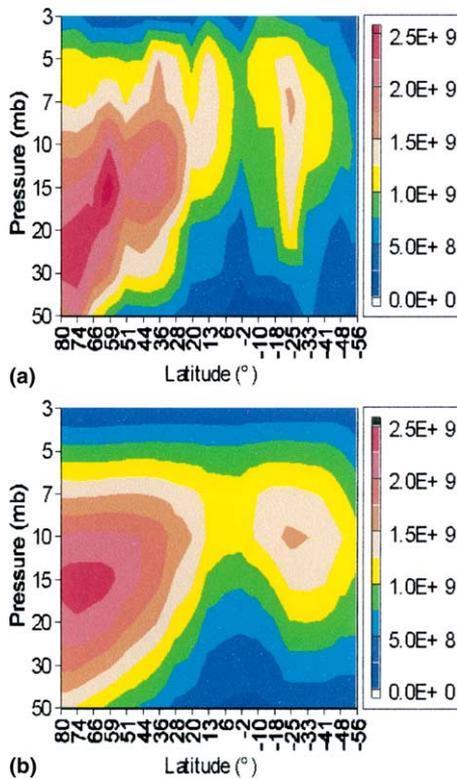


Fig. 4. (a) SCIAMACHY NO₂ number density (mol/cm³) on August 5, 2002. (b) Canadian Middle Atmosphere Model (de Grandpré et al., 1997) NO₂ meridional cross-section (mol/cm³) for the same conditions.

observations with our understanding of stratospheric photochemistry and dynamics (Fig. 4). The CMAM solar zenith angle is matched to that of SCIAMACHY by selecting the appropriate longitude from the global 3-D NO₂ snapshot. Longitudinal invariance within the model is thus assumed.

SCIAMACHY will provide unique coverage of lower stratospheric denoxification in polar winter. In Fig. 5, an example of this capability is illustrated. Notice

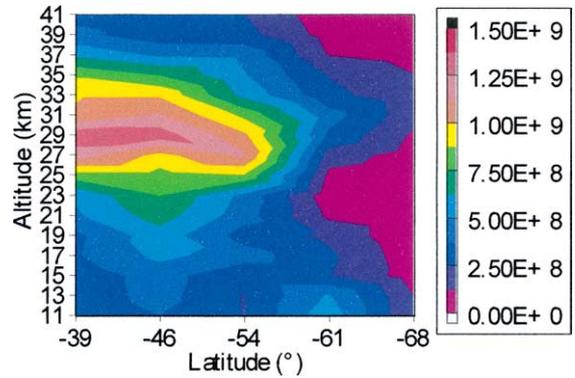


Fig. 5. Vertical and latitudinal distribution of NO₂ (mol/cm³) across the Antarctic vortex on 23/08/2002 (orbit 2508), retrieved using 435–495 nm window.

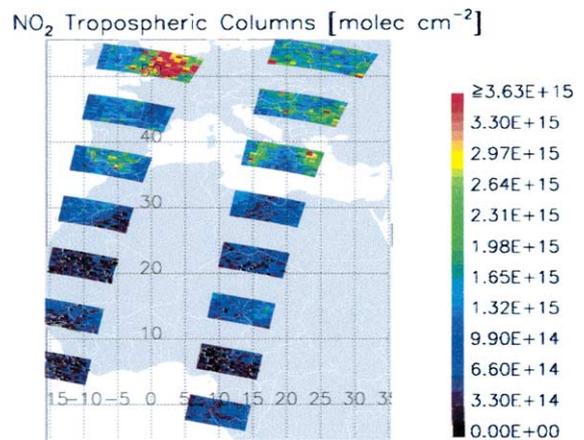


Fig. 6. Tropospheric residual from limb–nadir matching from SCIAMACHY on Friday, August 23, 2002 at 10 am LT.

that the denoxification does not increase monotonically downwards to the tropopause. At 61 and 68°S, the denoxified layer extends down to 19 and 14 km, respectively.

Fig. 6 shows the first retrieval of tropospheric NO₂ from limb–nadir matching. Thus far, the highest value, (8.55 ± 0.36) e16 cm⁻², has been observed over Los Angeles. The precision does not include the uncertainty of the air mass factor.

5. Conclusions

The “305 nm spectral knee technique” (S2003) often provides better tangent height information than either of the current satellite limb scatter instruments (i.e., SCIAMACHY and OSIRIS). Comparisons become favorable between these two instruments only after the TH correction is applied. A shift in TH does not translate into a simple shift in the altitude of the profile where the retrieval is non-linear due to optically thick Rayleigh scattering. This transition from optically thin to thick occurs at ~22 km but is SZA and wavelength

dependent. A comparison with HALOE, whose altitude registration is accurate to 150 m, also confirms the importance of an independent pointing determination. In summary, the preliminary agreement between SCIAMACHY and the correlative measurements is <10% in the 19–31 km range but increases to ~30% near 40 km.

Other satellite-borne instruments currently sensing profiles of NO₂ from its UV/visible absorption include SAGE II and III, POAM III, and OSIRIS. None of these instruments measure continuously over the optimized fitting window: 434.6–494.9 nm. Thus, all of these other instruments are at a disadvantage at sensing lower stratospheric NO₂. Also, a high sun is a second advantage in favour of SCIAMACHY over the other NO₂ profiling satellite instruments. This assists in the penetration of the lower stratosphere on the descending phase of the orbit. The extended fitting window also improves the penetration of the lowermost stratosphere by ~4 km. The extending fitting window is crucial for tropospheric NO₂ column retrievals because significant concentrations of NO₂ can reside in the lowermost stratosphere especially in the summer hemisphere at high latitudes as shown in Fig. 4(a).

Bovensmann et al. (1999) quoted precisions (based on spectral fitting) for the total nadir column and tropospheric column of 2% and 10%, respectively. The stratospheric component of the NO₂ column can usually be measured from limb scatter with 2% precision by integration of the vertical profile. The precision objective for the tropospheric NO₂ column was easily achieved in the metropolitan Los Angeles area. The profile precision from limb scattering of 10% between 20–40 km (Bovensmann et al., 1999) is also already achievable even on a 2 km grid. However, the theoretical precision limit of <15% between 10–40 km (Kaiser et al., 2002) has not been achieved.

One of the major sources of error in retrieving NO₂ profiles and tropospheric columns are the NO₂ cross sections. A ~205 K Fourier transform NO₂ absorption cross section spectrum accurate to 1% with a resolution of better than 10 cm⁻¹ is urgently required for accurate remote sensing, particularly for profiles of NO₂ in the upper troposphere and lower stratosphere, and for tropospheric residual columns from limb–nadir matching. This level of accuracy is also required at higher stratospheric temperatures.

The comparisons with OSIRIS and HALOE provide an early validation of SCIAMACHY NO₂ profiles at high latitudes in summer.

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