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Measurements of tropospheric NO_2 with an airborne multi-axis DOAS instrument

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Abstract

The AMAXDOAS instrument is an airborne multi-axis DOAS instrument covering the spectral range from 300 to 600 nm. During one flight of the SCIAVALUE campaign on 19 March 2003, the AMAXDOAS onboard the DLR Falcon detected tropospheric NO_2 over Europe under both cloudy and cloud free conditions. By combining the measurements in nadir and zenith direction, and analysing the spectra in the UV and the visible spectral region, information was derived on where the bulk of the observed NO_2 was located. Vertical columns of up to 5.7×10^{16} molec cm^{-2} were observed close to Frankfurt, in good agreement with surface measurements of 16.4 ppb. On several occasions, strong tropospheric NO_2 signals were also detected when flying above clouds. The ratio of zenith and nadir measurements indicates that the NO_2 observed was located within the cloud, and assuming the same profile as for the cloud free situation the NO_2 vertical column was estimated to be 5.0×10^{16} molec cm^{-2}. The results are relevant for the retrieval of tropospheric NO_2 columns from space-borne instruments in cloudy situations.

1. Introduction

Nitrogen dioxide, NO_2, is one of the most important trace gases in the atmosphere. In the troposphere its photolysis leads directly to the formation of O_3. High concentrations of NO_2 are harmful to vegetation and human health. Sources of NOx (NO_2+NO) include both natural (soils, lightning) and anthropogenic (fires, fossil fuel consumption) emissions.

In-situ measurements of NO_2 in the troposphere have been made by a variety of detectors, both on the surface and from air-borne platforms. In the last few years, retrievals of the tropospheric amount of NO_2 from GOME satellite measurements have yielded unprecedented information by providing a global view (e.g. Burrows et al., 1999; Leue et al., 2001; Richter and Burrows , 2002; Martin et al., 2002). Since August 2002,

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similar measurements are performed by the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) instrument on board of ENVISAT (Bovensmann et al., 1999) with improved spatial resolution. Although some successful first attempts have been made to validate the tropospheric NO_2 retrievals from space instrumentation (Heland et al., 2002, Heue et al., submitted, 2004[1]; Martin et al., submitted, 2004[2]), these have been limited to selected regions and many open questions remain. For the validation of the SCIAMACHY, two large campaigns were performed with the German research aircraft Falcon during September 2002 and in February and March 2003 (Fix et al., submitted, 2004[3]) . In this study, measurements taken by the Airborne Multi Axis DOAS (AMAXDOAS) instrument on board the Falcon on the 19 March 2003 have been analysed for NO_2, both in the presence and absence of cloud. The data have then been inverted, taking into account the dependence of the slant column on wavelength and viewing direction to retrieve the amount of NO_2 in the planetary boundary layer. Finally, conclusions are drawn for satellite validation and the interpretation of GOME and SCIAMACHY measurements.

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2. Methodology

2.1. Instrument

Airborne UV/visible spectrometers have been used in the past for studies of stratospheric (e.g Pfeilsticker and Platt, 1994; Petritoli et al., 2002) and tropospheric (McElroy et al., 1999; Melamed et al., 2003) composition. The AMAXDOAS instrument (Wagner et al., 2001) was specifically designed and constructed to retrieve tropospheric and stratospheric column amounts of atmospheric trace species for the validation of the SCIAMACHY instrument from aircraft. It comprises two grating spectrometers, one operating in the UV between 300 and 440 nm, the other covering the visible part of the spectrum (400–550 nm). Quartz fibre bundles are used to collect scattered sunlight from two sets of telescopes, one on the top and one on the bottom of the aircraft. During the flight discussed here, measurements were performed in four viewing directions: 0°, 88°, 92° and 180°, where the zenith direction is 180° and the nadir direction is 0°. The signals from the four directions are detected simultaneously with CCD imaging detectors: the data being averaged and then recorded about every 30 s, corresponding to 6 km spatial resolution along the flight track. The field of view of the AMAXDOAS is about 0.2°.

Analysis of the measurements is based on the well known Differential Optical Absorption Spectroscopy (DOAS) technique (Platt, 1994) and similar to that used for ground-based and satellite measurements. As has been shown in a recent sensitivity study, measurements from the different viewing directions of the instrument can be used to derive vertically resolved profiles (Bruns et al., 2004). Using a different approach, in this paper measurements from the zenith and nadir directions taken in different spectral regions having different weighting functions are combined to derive both the tropospheric NO_2 amount and its vertical distribution.

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2.2. Measurement situation

On the 19March 2003, the Falcon flight track was from Oberpfaffenhofen (48.1° N, 11.3° E, close to Munich, Germany, at 07:30 UT), to Denmark (at 09:00 UT), to the Netherlands, then to Belgium (at 10:00 UT) and back to Oberpfaffenhofen (at 10:48 UT). After take off the Falcon typically cruised at 11 km altitude. The flight track is shown in Fig. 1 on top of the cloud optical depth derived from MODIS data. As can be seen, the northern part of the flight was over a continuous bank of clouds, while the southern part was cloud free. According to the MODIS cloud top height data (King et al., 1997), the clouds above northern Germany were at about 700 hPa, while over the Netherlands and at the edge of the cloud field they were as low as 900 hPa. Local weather observations report that the cloud base was at about 400 m. It was a relatively warm day and the earth's surface was snow free under the flight track.

2.3. Sensitivity studies

To investigate the sensitivity of the measurements to NO_2 absorption in different altitudes below the aircraft, a number of model simulations were made using the radiative transfer model SCIATRAN (Rozanov et al., 1998, 2004). Calculations were performed at three representative wavelengths (360, 433, 485 nm) where NO_2 retrieval is possible for both the nadir and the zenith viewing geometry.

Two specific cases were distinguished: a) a cloud free situation and, b) a measurement above a low cloud. For the simulations, temperature and pressure profiles were taken from the US standard atmosphere, and an urban aerosol with a visibility of 10 km was assumed in the boundary layer. The flight altitude was set to 11 km, and the solar zenith angle (SZA) was selected to be 60°. The surface spectral reflectance was set to 0.02, 0.05 and 0.08 at the wavelengths 360, 433, and 485 nm, respectively. To test the albedo effect, a scenario with a high albedo (0.8) was also investigated.

For the simulations with cloud, a cloud layer between 0.4 and 1.5 km was assumed with an optical depth of 25, which is about the average cloud optical depth observed

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by MODIS along the flight track. As the instrument field of view is small and the clouds were continuous and large, partially cloudy scenes were not considered.

In Fig. 2 (left), the weighting functions for the clear sky case and low albedo are shown at the three wavelengths in both nadir and zenith direction. The weighting functions describe the change of the measured NO_2 column for a change in concentration in a given altitude relative to the corresponding change in the vertical column. As can be seen, for small albedo the sensitivity to the boundary layer differs significantly between the three wavelengths, whereas the relative differences are small in the free troposphere. This is predominantly a result of increased Rayleigh scattering in the UV which reduces the penetration depth of the photons and thereby the sensitivity to the lowest layers, but the change in surface reflectivity also contributes. By analysing the ratio of measurements at different wavelengths, an estimate can be made on where the bulk of the NO_2 is located and at least boundary layer and free tropospheric NO_2 should be distinguishable. At a given wavelength, the ratio of nadir and zenith weighting function is also decreasing with altitude, and depending on the measurement uncertainty, this information could also be used to derive altitude information.

As shown in Fig. 2 (middle) the weighting functions for high albedo (0.8) are much larger in the boundary layer than those for small albdeo and also the decrease with altitude is smaller. As the relative differences between the different wavelengths decrease, the amount of profile information is much lower for such a situation which occurs for example over snow or fog.

In the presence of a cloud, the sensitivity of the measurements to NO_2 below the cloud is close to zero. Within the cloud, the sensitivity increases and above the cloud, it is larger than for the case without cloud (Fig. 2, right). This is the result of the higher effective spectral reflectance of the cloud top and the multiple scattering within the cloud. In contrast to the cloud free situation, the weighting functions for the three wavelengths do not differ significantly. The sensitivity of the zenith direction is again much smaller than that of the nadir direction, but above the cloud, it is also larger than in the cloud free situation. These relationships do not depend significantly on the cloud's

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altitude as long as the cloud is below the aircraft. The cloud optical depth determines the gradient of the weighting functions within the cloud and also the absolute value above the cloud. Surface albedo plays no role for an optically thick cloud.

According to the figure, a small signal should be observed in the zenith direction if there is NO_2 above the cloud or in its upper part, but not if it is within or below the cloud. As the nadir weighting function decreases much slower within the cloud, the ratio between the measurements in nadir and zenith direction can be used to estimate if NO_2 is above, below or within the cloud.

3. Results and discussion

The AMAXDOAS measurements from 19 March 2003 were analysed in three different wavelength windows, 345–380 nm, 410–456 nm, and 472–497 nm, respectively. As the focus of these measurements are the tropospheric concentrations, a measurement taken at about 09:36 UT, selected to be in a clean region free from significant tropospheric NO_2 has been used as a background spectrum. The stratospheric signal is assumed to be constant over the next hour of measurement. The resulting columns are therefore differential slant columns relative to that in the background measurement. To analyse the tropospheric NO_2 column, the NO_2 cross section at 273 K was used in the retrieval.

In Fig. 3, the NO_2 slant columns retrieved from the nadir and zenith viewing directions in the 410–456 nm window are shown with the O_4 slant columns derived in the 345–380 nm window nadir viewing direction. The O_4 columns are clearly enhanced in some regions, indicating low cloud, in agreement with the MODIS cloud data shown in Fig. 1 and the observations during the flight. Large nadir columns are retrieved in the clear region at the end of the flight, close to Frankfurt (at about 10:30 UT). However, on several occasions, enhancements of NO_2 were also observed in cloudy situations, in particular when approaching cloud free areas, for example at 09:48 UT between Amsterdam and Utrecht. NO_2 columns in the zenith direction (see Fig. 3) are small

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throughout the flight, but tend to increase when nadir NO_2 is large.

As the vertical profile of the oxygen dimmer, O_4, is known, and ignoring small effects resulting from surface pressure variability, the effective light path of the measurements can be estimated from the slant column change. The O_4 slant column depends mainly on surface elevation, albedo and cloud. High clouds will reduce the observed column as they hide part of the column while low clouds increase the column through the effect of increased albedo and multiple scattering. For the measurements having low cloud below the Falcon, the cloud top height and optical thickness of the cloud were derived from the O_4 measurements. This was achieved by varying the cloud parameters used in the radiative transfer calculations until the derived O_4 vertical columns agreed for nadir and zenith measurements as well as in cloudy and clear sky conditions. Good consistency was obtained for a low cloud layer between 0.4 and 1.5 km having an optical depth of 25 as illustrated in Fig. 4. These numbers are also consistent with the MODIS data (see Fig. 1). Therefore these cloud settings were used for the evaluation of the NO_2 columns in the cloudy situation, and the weighting functions shown in Fig. 2 already used these settings.

As discussed above, the sensitivity of the retrieval to the boundary layer NO_2 depends on wavelength. This effect is demonstrated in Fig. 5, where NO_2 slant columns retrieved in the three different wavelength regions (345–380 nm, 410–456 nm, and 472–497 nm) are shown. NO_2 columns at all wavelengths are very similar over clouds, showing the high degree of consistency of the AMAXDOAS NO_2 measurements. Under cloud free conditions, the enhanced columns retrieved from the three fitting windows differ significantly and increase with wavelength. The ratio of the slant columns retrieved at 410–456 nm and 345–380 nm is about 2. According to the sensitivity studies presented above, this indicates that the bulk of the NO_2 must be located in the boundary layer, probably resulting from local emissions.

As the sensitivity of the measurements to NO_2 below the clouds is negligible, the NO_2 signal observed over the cloud must originate from NO_2 above or in the uppermost part of the cloud. As the zenith viewing columns also show some enhancement in these

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regions (see Fig. 3), it is concluded that significant amounts of NO_2 must have been above or in the cloud top. Whether or not there is NO_2 below the clouds can not be determined from the measurements due to the very low sensitivity.

As can be seen from the cloud weighting function the ratio of nadir and zenith NO_2 column should be between 2 and 3 if the NO_2 layer is above the clouds. If the cloud layer is between 1.5 and 2.0 km, the ratio of nadir and zenith NO_2 column as simulated with the radiative transfer model is 2.7. However, in the measurements, the ratio of the nadir and zenith NO_2 slant column is about 4.8, indicating that the NO_2 is located in the upper part of the cloud and not as a layer above the cloud.

The measured NO_2 columns in three wavelength windows were converted to vertical columns with air mass factors (AMFs) based on the above described scenarios. The AMF is defined as the ratio between slant column and vertical column of the absorber. For optically thin constituents it can be calculated from the product of weighting function and concentration profile. According to the temperature and wind measurements on the Falcon during the descent at about 10:48 UT, the boundary layer height was about 1.3 km. We therefore assume that NO_2 is well mixed below 1.3 km and linearly decreases to 0 at 1.6 km in both the cloudy and cloud free situation. The NO_2 vertical columns are shown in Fig. 6 for the last part of the flight including the large peak over the cloud. In the cloudy situation, the NO_2 column was about 5.0×10^{16} molec cm^{-2} and about 5.7×10^{16} molec cm^{-2} in the cloud free peak. The agreement of the NO_2 vertical columns in the three wavelength regions gives a consistent picture of enhanced NO_2 in the boundary layer.

The NO_2 vertical columns retrieved from the AMAXDOAS measurements were compared with ground-based in-situ measurements from the Hessisches Landesamt für Umwelt and Geologie (http://www.hlug.de/medien/luft/index.htm). The in-situ NO_2 measurement at 10:30 UT was selected from seven air quality measurement stations within 20 km of the flight track. The average of these measurements yields 32.6 μg m^{-3}, which corresponds to about 16.4 ppb. When converted to a vertical column using the profile assumed for the AMF calculation, this corresponds to a vertical column

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of 5.5×10^{16} molec cm^{-2}, which is consistent with the AMAXDOAS measurements of 5.7×10^{16} molec cm^{-2}.

The NO_2 columns, observed over the cloud close to Amsterdam are comparable to those observed in the cloud free regions over polluted sites. It is therefore reasonable to assume that they are the result of either vertical transport through the cloud, or more for low clouds, the uplifting of BL air in the cloud. As the position of the enhanced NO_2 values over the cloud is mostly close to cloud edges in the data set, horizontal transport related to the movement of the cloud fields could also play a role.

4. Conclusions

AMAXDOAS measurements of scattered light from a flight over Denmark, the Netherlands, Belgium and Germany on 19 March 2003 are reported. Tropospheric NO_2 columns were retrieved from the measurements and strongly enhanced values were observed under clear sky conditions close to Frankfurt, but also over clouds, in particular close to the cloud edge in polluted regions.

Sensitivity studies using the radiative transfer model SCIATRAN show that for clear sky conditions, the sensitivity to boundary layer NO_2 changes by more than a factor of 2 between 360 and 485 nm. Analysis of the NO_2 slant column in three different wavelength windows therefore yields information about the vertical location of the NO_2. In the case studied here, the results from three wavelength regions were shown to be consistent with NO_2 being located in a boundary layer of 1.3 km height. This is the first demonstration of vertically resolved retrieval of NO_2 from measurements aboard a high altitude aircraft. The tropospheric NO_2 vertical columns derived in cloud free situation were compared with ground based in-situ measurements, and good agreement was found.

For cloud covered situations, the cloud top height and an estimate of the cloud optical depth were derived from the observed O_4 columns, measured in nadir and zenith direction. For the flight discussed here, a cloud top height of 1.5 km and an optical

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depth of 25 were estimated from the measurements. These values are consistent with MODIS cloud data determined at about the same time.

Sensitivity studies for measurements over a low and optically thick cloud show that the vertical sensitivity depends only weakly on wavelength in agreement with the measurement results. For zenith sky measurements above clouds it could be shown that a small but significant signal originates from the light scattered upwards from the cloud and then back to the aircraft. The observation in zenith direction, coupled with the nadir measurements were used to determine whether enhanced NO_2 columns observed over clouds originated from NO_2 within or above the cloud. The measurements taken during this flight indicate that the NO_2 signal observed in cloudy situations were mainly from NO_2 in the upper part of the cloud but not above the cloud.

The results of this study have important implications for satellite measurements of tropospheric NO_2, for example from GOME or SCIAMACHY. First, the wavelength method described here is also applicable to satellite measurements, providing a new method to derive BL NO_2 from space (Richter and Burrows, 2000). Second, the observation of NO_2 in cloudy situation is an important constraint for attempts to estimate BL NO_2 from satellite measurements by comparing cloudy and cloud free measurements. And last, the observation of enhanced NO_2 over the edge of cloud fields indicates a potential problem for NO_2 retrieval from partially cloudy satellite scenes, as the relatively large NO_2 signal from the highly visible NO_2 in the upper part or above the cloud could be misinterpreted to be an inappropriately large amount of NO_2 in the BL.

The sensitivity studies and measurement results presented here demonstrate the large potential of air-borne UV/visible absorption measurements for studies of tropospheric NO_2 pollution. Such measurements are well suited for satellite validation and can also contribute to environmental and air quality monitoring.

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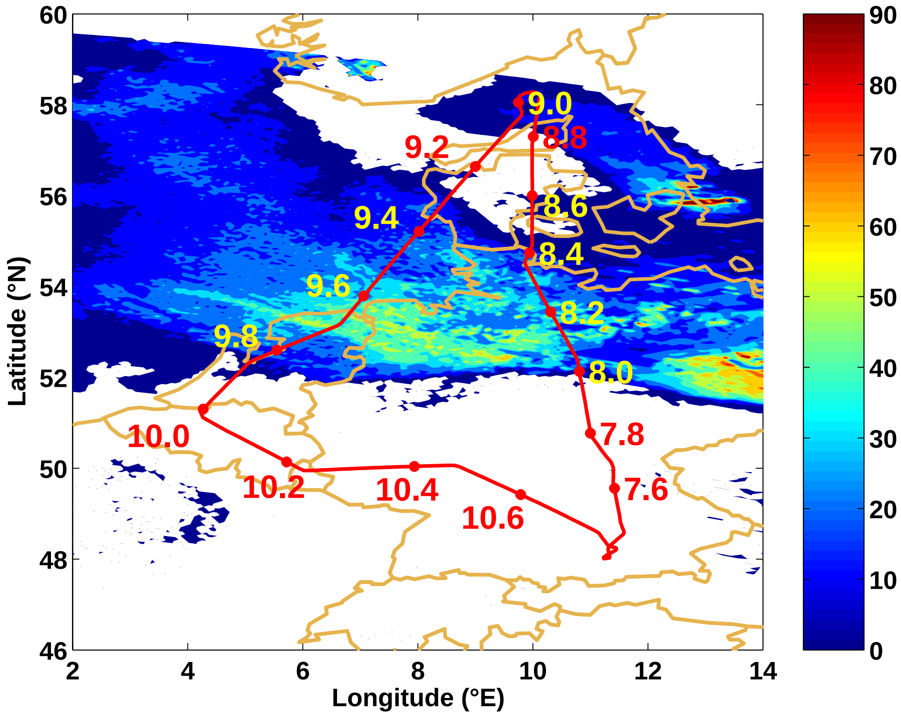

Fig. 1. MODIS cloud optical depth map for 19 March 2003, at 10:50 UT. The red line indicates the Falcon flight track with approximate times (UT). White areas are cloud free regions. The flight was from 07:30 UT to 10:48 UT with solar zenith angles ranging from 50° to 70°.

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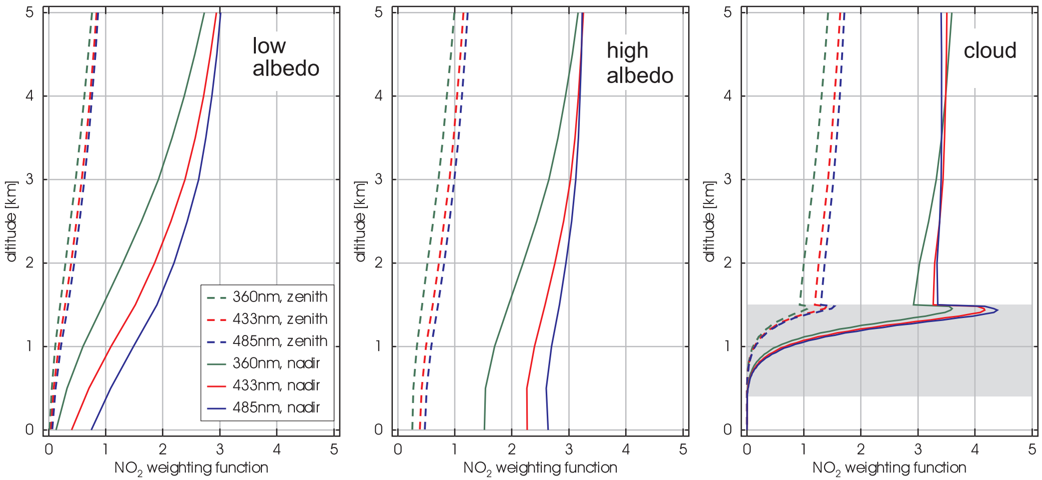

Fig. 2. Weighting functions or altitude dependent airmass factors for clear sky, low albedo (left), clear sky and high albedo (middle) and with a cloud (right). The shaded area indicates the position of the cloud. Solid lines are for the nadir viewing direction, the broken lines for zenith observation. All curves are for a flight altitude of 11 km.

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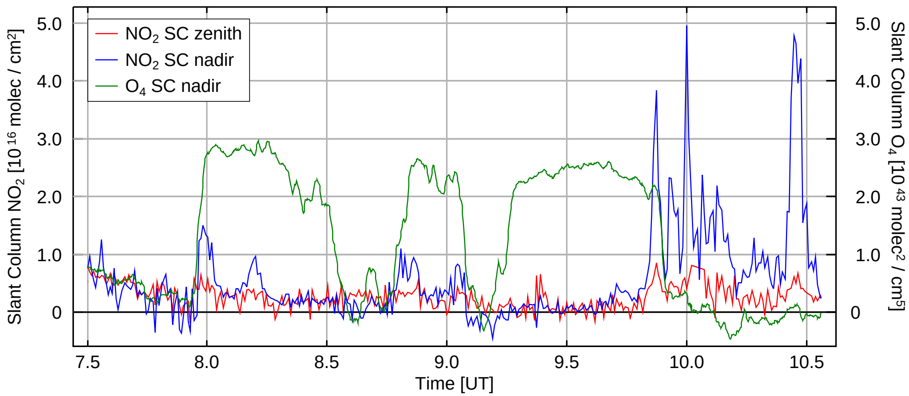

Fig. 3. NO_2 slant columns in zenith and nadir direction, and nadir O_4 columns observed during the Falcon flight. The solar zenith angle varied from 70° to 50°. The background spectra were taken at 09:36 UT at a solar zenith angle at about 61°.

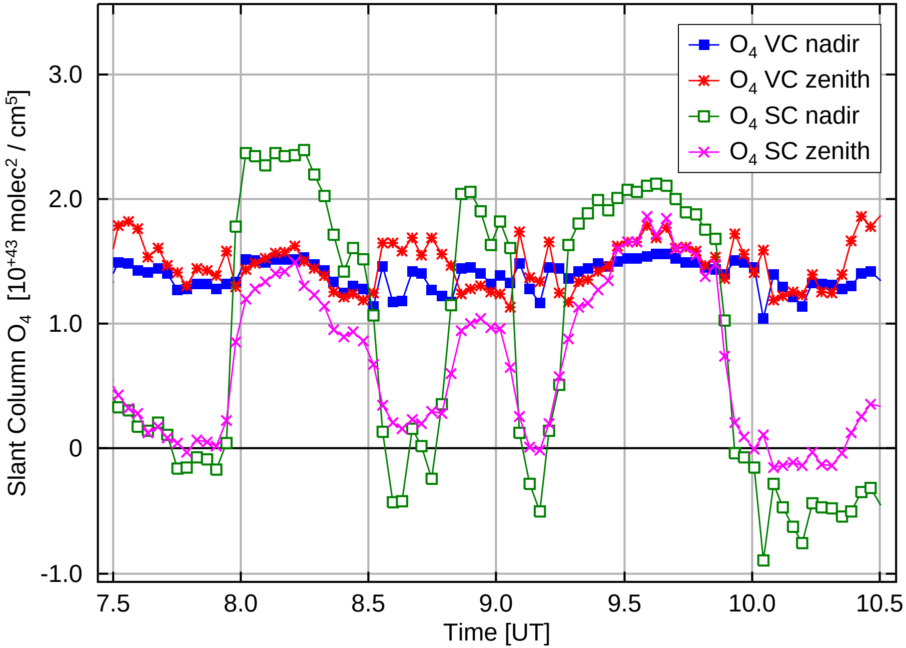

Fig. 4. O_4 slant and vertical columns observed in zenith and nadir direction during the flight. For the cloudy parts, a low cloud layer between 0.4 and 1.5 km with an optical depth of 25 was assumed in the airmass factor calculations.

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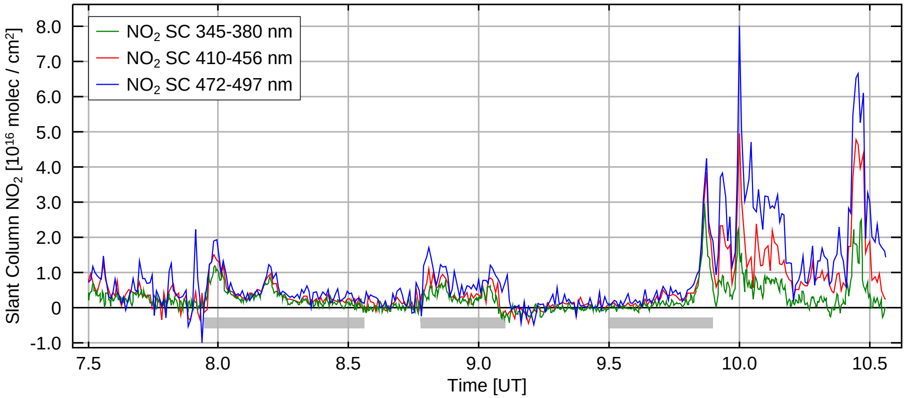

Fig. 5. NO_2 slant column retrieved in different windows (regions having cloud are indicated by the grey bars). Over polluted regions, measurements at larger wavelength yield larger slant columns in cloud free situations but not over clouds.

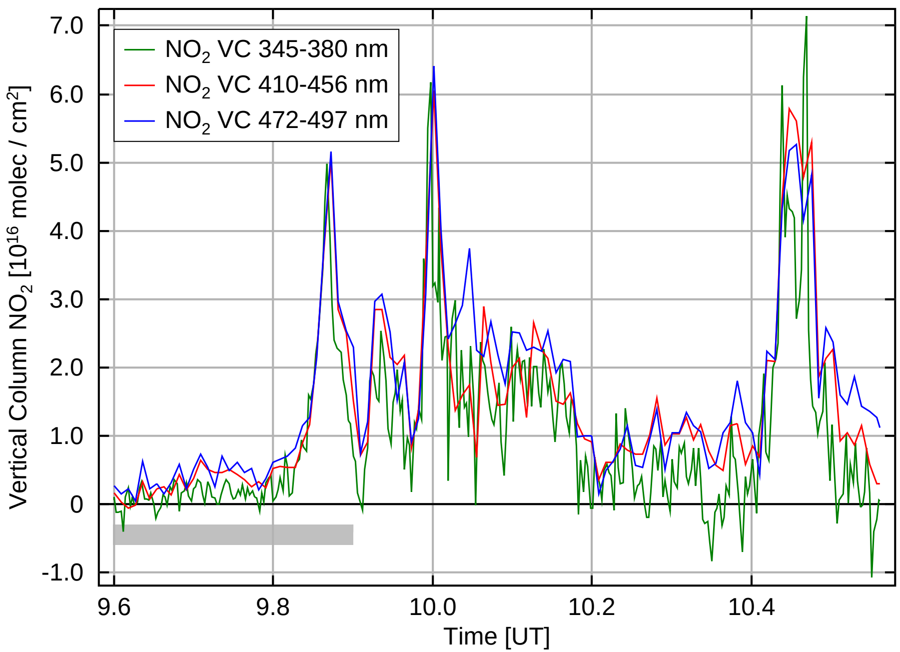

Fig. 6. NO_2 vertical columns for the Falcon flight calculated from different fitting windows using appropriate airmass factors with and without clouds.

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