

NO₂ Profile retrieval using airborne multi axis UV-visible skylight absorption measurements over central Europe

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Abstract. A recent development in ground-based remote sensing of atmospheric constituents by UV/visible absorption measurements of scattered light is the simultaneous use of several directions with small elevation angles in addition to the traditional zenith-sky pointing. The different light paths through the atmosphere enable the vertical distribution of some atmospheric absorbers such as NO₂, BrO or O₃ to be retrieved.

In this study, the amount of profile information that can be retrieved from such measurements on aircraft is investigated for the trace gas NO₂. A Sensitivity study on synthetic data is performed for a combination of four lines of sight (LOS) (0°(nadir), 88°, 92°, and 180° (zenith)) and three wavelength regions [center wavelengths: 362.5 nm, 437.5 nm, and 485.0 nm]. The method used in this work is a combination of two previously established methods described in Petritoli et al. (2002) and Wang et al. (2004). The investigation presented here demonstrates the potential of this LOS/wavelengths setup to retrieve a significant amount of profile information from airborne multi-axis differential optical absorption spectrometer (AMAXDOAS) measurements with a vertical resolution of 3.0 to 4.5 km in the lower troposphere and 2.0 to 3.5 km near flight altitude. Above 13 km the profile information content of AMAXDOAS measurements is sparse. The retrieval algorithm used in this work is the AMAXDOAS profile retrieval algorithm (APROVAL).

Further, retrieved profiles with a significant amount (up to 3.2 ppbv) of NO₂ in the boundary layer over the Po-valley (Italy) are presented. Airborne multi-axis measurements are thus a promising tool for atmospheric studies in the troposphere.

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1 Introduction

The airborne multi-axis differential optical absorption spectrometer (AMAXDOAS) is a remote sensing instrument built to detect a number of different trace gases such as O₃, NO₂, BrO, OClO, and SO₂. AMAXDOAS was used to validate (Heue et al., 2005) measurements of the scanning imaging absorption spectrometer for atmospheric cartography (SCIAMACHY) (Bovensmann et al., 1999). The former has been flown in two major campaigns (SCIAMACHY validation utilization experiment (SCIA-VALUE)) in September 2002 and February/March 2003 (Fix et al., 2005).

In the troposphere Nitrogen Dioxide (NO₂) is an important trace gas since its photochemistry is involved in the production of tropospheric Ozone. In densely populated areas the most important source of NO₂ are anthropogenic emissions. Thus the monitoring of NO₂ concentrations in these areas is necessary because Ozone and NO₂ itself are harmful species affecting both human health and the growth of vegetation. Since all NO₂ emissions affect the planetary boundary layer directly the monitoring of the NO₂ levels in the boundary layer is getting more and more important. We present a novel tool consisting of a remote sensing instrument and a profile retrieval method to accomplish this task.

To understand the NO_x chemistry in more detail an acquisition of the vertical distribution of trace gases is necessary. Thus an instrument able to resolve the vertical profile information of trace gases is important. The use of airborne UV/visible spectrometers to study the tropospheric (McElroy et al., 1999; Melamed et al., 2003) and stratospheric (Pfeilsticker et al., 1994; Petritoli et al., 2002) composition is a well known technique. In recent years different groups (Petritoli et al., 2002; Melamed et al., 2003) presented interesting results of limited profile information using the DOAS method and different LOS observed from an airborne platform. The latest study presents a profile retrieval for

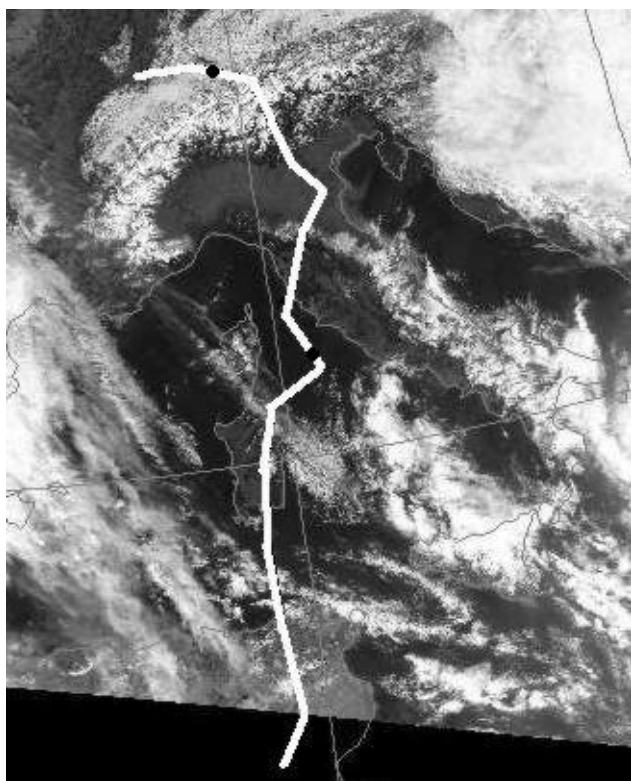


Fig. 1. Flight track of flight 030219 and meteorological situation. The satellite image is from the MODIS instrument (channel 4) onboard the TERRA satellite (source: <http://www.sat.dundee.ac.uk>). The data was acquired on 19 February 2003 at 10:26 UTC. Results are presented in this paper from the section of track between the black dots.

Ozone using airborne multiaxis UV/visible measurements (Liu et al., 2005).

Here, we have used several lines of sight (LOS) in combination with several wavelength regions (4-3 setup) to maximize the content of profile information. The careful selection of the LOS and wavelength regions enables the retrieval of a significant amount of profile information from the AMAXDOAS measurements. Compared to the profile retrieval method using only a four LOS setup as presented in Bruns (2004) the 4-3 setup improves the retrieved profiles significantly. This new setup demonstrates significant improvements even compared to some LOS setups using ten LOS but only one wavelength region as shown in Bruns et al. (2004). A different method to derive vertical distributions for trace gases from AMAXDOAS data was used by Wang et al. (2004). Wang et al. (2004) are using only the nadir and zenith LOS at three different wavelength regions. Their method enables the determination of boundary layer NO₂ only with the use of significant a priori information regarding the height of the planetary boundary layer and the profile shape. The profile retrieval method described in this work does not rely on any a priori information regarding the tro-

posphere (i.e. boundary layer height and/or profile shape) since the a priori profile shows no NO₂ in the troposphere. The only a priori information the presented profile retrieval method uses is regarding the stratospheric NO₂ profile.

2 Experimental

2.1 Instrument

The AMAXDOAS instrument was designed to detect atmospheric abundances of different trace gases like Ozone, NO₂, BrO, OClO, SO₂, and Formaldehyde. It consists of two grating spectrometers, one operating in the UV wavelength region (300 to 440 nm) and the other operating in the visible wavelength region (400 to 550 nm). The scattered skylight is collected by several telescopes (one per spectrometer and LOS) and directed into the spectrometers using a quartz fiber bundle. The spectral information is recorded by two CCD-detectors and passed on to two computers for data storage. A more detailed description of the AMAXDOAS experimental setup can be found in Bruns (2004). The AMAXDOAS instrument simultaneously measures in four different LOS (0°(nadir), 88°, 92°, and 180°(zenith)). The different measurement geometries resulting from this technique are described in Bruns et al. (2004); Bruns (2004). The novelty of the profile retrieval method described in this investigation is the combination of four LOS with three different wavelength regions (362.5, 437.5, and 485 nm) resulting in twelve virtual LOS. The additional information from the different wavelength regions improves the vertical resolution of the retrieved profiles significantly as can be seen in the sensitivity study below.

The well known differential optical absorption spectroscopy (DOAS) method was used to analyze the spectral information recorded by the AMAXDOAS instrument (Solomon et al., 1987; Platt, 1994).

2.2 Measurements

The measurements were gathered during two major campaigns (SCIA-VALUE) in September 2002 and February/March 2003 (Fix et al., 2005) in the context of validation of the SCIAMACHY instrument onboard ESA's ENVISAT. The flight routes for both campaigns were chosen to cover latitudes from the Arctic to the tropics as well as a significant longitudinal cross section.

In this study measurements from a flight on 19 February 2003 have been analyzed to retrieve profile information for the trace gas NO₂. This flight started in Basel, Switzerland, and headed for Tozeur, Tunisia, crossing the Alps and Italy with a flight altitude ≥ 10 km. Figure 1 shows the flight track and the meteorological situation for this flight. The MODIS satellite image acquired on the same day at 10:26 UTC shows no clouds for the first part of the flight. Over the Alps the cloud situation is hard to evaluate from Fig. 1 because of

the snow covered mountains. The observations made by the operator onboard the aircraft stated a cloud free situation over the Alps and haze over the Po valley.

3 The profile retrieval

3.1 Method

The profile retrieval method used in this work is the same as presented in Bruns et al. (2004); Bruns (2004) giving a detailed description. It is based on the Optimal Estimation Method described by Rodgers (2000). A set of measurements \mathbf{y} can be related to a vertical profile \mathbf{x} by a forward model F :

$$\mathbf{y} = F(\mathbf{x}, \mathbf{b}) + \epsilon \quad (1)$$

where \mathbf{b} is the vector of the forward model parameters and ϵ is the sum of the measurement error and the model error. In our case, \mathbf{y} is a vector of slant columns as a function of LOS and wavelength obtained from the AMAXDOAS raw spectra using the DOAS method, and \mathbf{x} is the vertical profile of the trace gas of interest. The profile \mathbf{x} – a continuous function in the real atmosphere – has to be sampled discretely by the retrieval algorithm and is therefore presented as a vector. Equation (1) can be rewritten in a linearized form:

$$\Delta \mathbf{y} = \mathbf{K} \Delta \mathbf{x} \quad (2)$$

where $\Delta \mathbf{x}$ is the perturbation in the vertical profile, $\Delta \mathbf{y}$ is the change in the slant columns due to the perturbation in the vertical profile, and

$$\mathbf{K} = \frac{d\mathbf{y}}{d\mathbf{x}} \quad (3)$$

The rows of the \mathbf{K} matrix represent the weighting functions, and each row corresponds to a different measurement taken at a specific LOS and in a specific wavelength region. The weighting functions characterize the sensitivity of the measured slant columns \mathbf{y} to the variation of the vertical profile \mathbf{x} . The forward model used in this study to calculate the weighting functions is the radiative transfer model SCIATRAN (Rozanov et al., 2001).

SCIATRAN calculates the weighting functions by solving the linearized radiative transfer equation. For a more detailed description of how SCIATRAN actually calculates weighting functions see Rozanov et al. (1998).

In this study the Maximum a Posteriori (MAP) solution is chosen (Rodgers, 2000). This method calculates the retrieved profile as follows:

$$\hat{\mathbf{x}} = \left(\mathbf{K}^T \mathbf{S}_\epsilon^{-1} \mathbf{K} + \mathbf{S}_a^{-1} \right)^{-1} \left(\mathbf{K}^T \mathbf{S}_\epsilon^{-1} \mathbf{y} + \mathbf{S}_a^{-1} \mathbf{x}_a \right) \quad (4)$$

where \mathbf{K} is the weighting function matrix, \mathbf{S}_ϵ is measurement error covariance matrix, \mathbf{S}_a is the error covariance matrix of

the a priori error, \mathbf{y} is the measurement vector, and \mathbf{x}_a is the a priori profile information.

To characterize the retrieved profile more precisely, the averaging kernel matrix \mathbf{A} is introduced:

$$\mathbf{A} = \mathbf{D} \mathbf{K}; \quad \mathbf{D} \equiv \mathbf{S}_a \mathbf{K}^T \left(\mathbf{K} \mathbf{S}_a \mathbf{K}^T + \mathbf{S}_\epsilon \right)^{-1} \quad (5)$$

where \mathbf{D} are the so-called contribution functions. The averaging kernel describes the sensitivity of each layer of the retrieved profile on the variation of the true vertical profile.

3.2 Error analysis

The total error of the retrieved profile can be separated into three components. According to Rodgers (2000) the total error of the profile retrieval is the difference between the retrieved and the true profile. According to error propagation the error covariance matrix of the total error can be written as:

$$\mathbf{S}_{\text{tot}} = \mathbf{S}_n + \mathbf{S}_m + \mathbf{S}_f \quad (6)$$

\mathbf{S}_n is the smoothing error covariance matrix, \mathbf{S}_m is the retrieval noise covariance matrix, and \mathbf{S}_f is the forward model error covariance matrix. The last error component will not be considered in this work because the error produced by the forward model SCIATRAN is less than 2% (neglecting the uncertainty of the aerosol profile optical properties) for LOS with tangent heights up to 30 km (Rozanov et al., 2001).

The smoothing error covariance matrix \mathbf{S}_n can be calculated as:

$$\mathbf{S}_n = \left(\mathbf{A} - \mathbf{I} \right) \mathbf{S}_a \left(\mathbf{A} - \mathbf{I} \right)^T \quad (7)$$

where \mathbf{S}_a is the error covariance matrix of the a priori profile. The diagonal elements of \mathbf{S}_a have been determined empirically. The unit of the diagonal elements of \mathbf{S}_a is ppbv². In other words the set of diagonal elements of \mathbf{S}_a resulting in profiles having the smoothest shape was chosen. The following values for the elements of \mathbf{S}_a have been used for the retrieval grid (1 km, 4 km, 7 km, 9 km, 11 km, 13 km, 39 km): 2.8, 0.4, 0.1, 0.1, 0.1, 0.1, and 0.01 above 13 km altitude. \mathbf{S}_a also contains extra-diagonal elements to take into account correlations between NO₂ values at different altitude levels. The extra diagonal elements of \mathbf{S}_a are calculated using a Gaussian function as follows (Barret et al., 2002):

$$\mathbf{S}_{a_{ij}} = \sqrt{\mathbf{S}_{a_{ii}} \mathbf{S}_{a_{jj}}} \exp\left(-\ln(2) \left((z_i - z_j) / \gamma \right)^2\right) \quad (8)$$

where z_i and z_j are the altitudes of levels i and j . γ is the correlation length represented as half width at half maximum (HWHM). In this work γ was set to 1.5 km which translates to a correlation length of 3 km. This choice was made because the largest step size of the retrieval grid points is 3 km.

Rodgers (2000) refers to \mathbf{S}_n as smoothing error covariance matrix, due to the fact that this covariance matrix corresponds to portions of profile space the measurements cannot

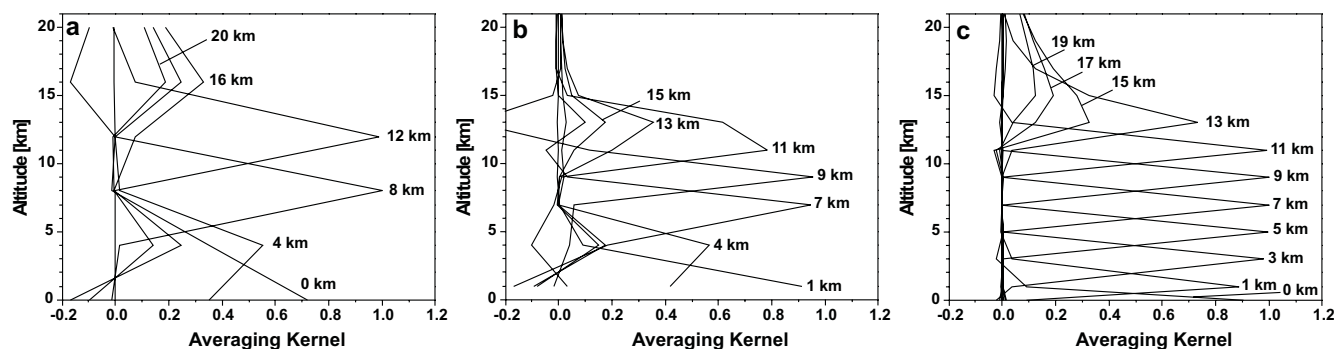


Fig. 2. Averaging kernels of different line-of-sight setups: LOS at 500 nm (a), four LOS and three wavelengths (b), and 18 LOS at 500 nm (c).

Table 1. Urban aerosol setting.

layer	rel. humidity [%]	aerosol component	relative mixing ratio
0–2 km	80	water soluble	0.31399
		insoluble (dust)	0.00001
		soot	0.68600
2–10 km	70	water soluble	0.45790
		insoluble (dust)	0.00010
10–30 km	0	soot	0.54200
30–60 km	0	sulfate	1.00000
		meteoric dust	1.00000

see. In our case those portions are the higher altitudes in the stratosphere, and small scale variations obscured by the limited altitude resolution of the profile retrieval.

The retrieval noise covariance matrix S_m can be calculated as:

$$S_m = DS_\epsilon D^T \quad (9)$$

where S_ϵ is the covariance matrix of the measurement error and D is the contribution function matrix. This error component is caused by noise in the measurements propagating into the retrieval. The contribution function matrix maps the measurement error into the profile space since the dimension of the measurement error covariance matrix is different compared to the dimension of the a priori covariance matrix and Eq. (6) is used to add up all error components to result in a total retrieval error. S_ϵ is a diagonal matrix with the diagonal elements being the squares of the individual measurement errors of each LOS.

4 Sensitivity study

Before discussing the results involving real AMAXDOAS data a sensitivity study dealing with the four LOS and three

wavelengths setup (4-3 setup) is presented. The 4-3 setup is compared to two other setups to evaluate the quality of the retrieved profiles using the 4-3 setup. This study is intended to demonstrate the potential of the 4-3 setup to retrieve profile information since this retrieval method using different LOS in combination with three different wavelength regions is new. The general assumptions made for this sensitivity study are:

- flight altitude 10 km
- surface albedo of 0.1
- solar zenith angle (SZA) is 51.6°
- no clouds
- trace gas NO₂
- retrieval grid: 1, 4, 7, 9, 11, ..., 39 km
- a priori error of 4 ppbv at 1 and 4 km, 1 ppbv otherwise
- measurement error of 10¹⁵ molec/cm²
- urban aerosol profile (see Table–1 and Fig. 4)

Figure 2 shows the averaging kernels of three different LOS setups. Figure 2a represents the averaging kernels of a 4-1 setup including the LOS 0° (nadir), 88°, 92°, and 180° (zenith) at 500 nm. As can be seen the retrieved profile information is very limited due to the poor vertical resolution of about 4.0 km. Figure 2b depicts the averaging kernels for the 4-3 setup (four LOS: 0° (nadir), 88°, 92°, and 180° (zenith) at three different wavelength regions: 362.5 nm, 437.5 nm, and 485.0 nm). The retrieved profiles using this setup contain significantly more profile information than the retrieved profiles using only four LOS. Figure 2c shows the averaging kernels for a 18-1 setup with 18 LOS (0°, 80°, 85°, 86.8°, 87.0°, 87.3°, 87.7°, 88.2°, 89.0°, 91.0°, 91.8°, 92.3°, 92.7°, 93.0°, 93.2°, 95°, 100°, and 180°) at 500 nm. The advantage of using so many LOS is the perfect scanning of the atmosphere in limb mode meaning the LOS 86.8°, 87.0°, 87.3°,

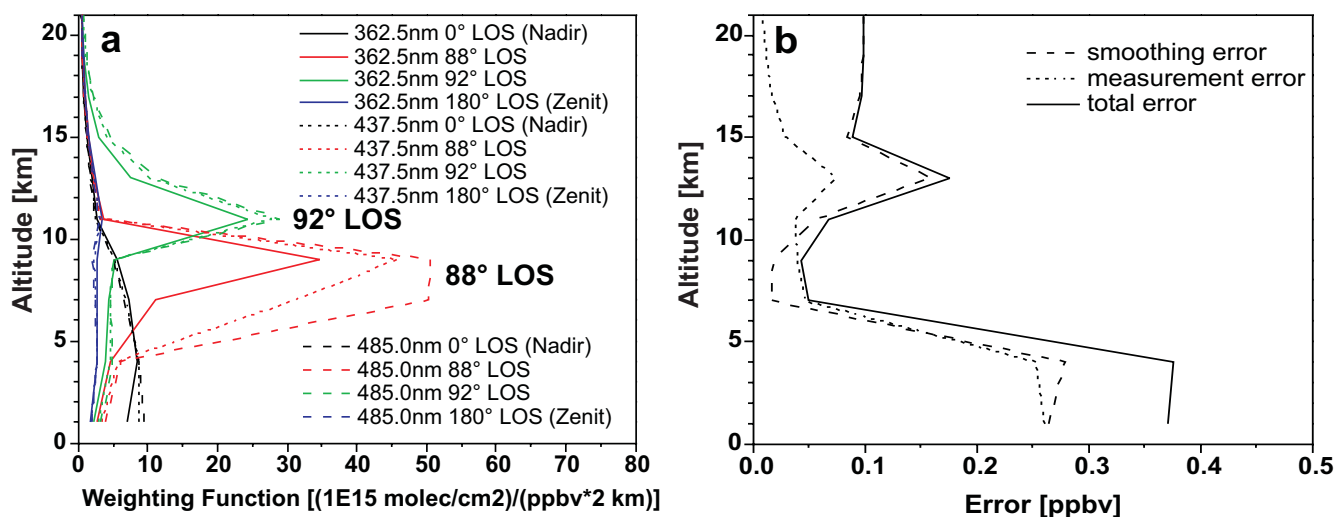


Fig. 3. Weighting functions (a) and errors (b) for the combination of four LOS and three wavelengths.

87.7°, 88.2°, and 89.0° translate to tangent heights of 0 km, 1 km, 3 km, 5 km, 7 km, and 9 km respectively. It can be seen in this plot that the profile information has increased significantly compared to the four LOS and 4-3 setups because of the very good vertical resolution of close to 2.0 km. And there is still potential to improve the vertical resolution down to 1.0 km when using a 18-3 setup (18 LOS at three wavelength regions). The main reason the number of different LOS was set to four is the increase of the signal-to-noise ratio (SNR). Since all LOS are recorded simultaneously on the CCD-chip fewer LOS result in more lines of the CCD-chip per LOS. The increased SNR produces smaller retrieval errors. Tests with a 10-1 setup have shown that the SNR is too small to get good results from the measured data. Another problem to be solved is that the viewing directions must be determined to an accuracy of much smaller than 0.1° when using so many LOS pointing almost in the same direction.

The 4-3 setup proved to be the optimum of LOS setups for the AMAXDOAS instrument considering the boundary conditions mentioned above. Figure 3 shows the results of a profile retrieval using the 4-3 setup. Plot a) presents the weighting functions indicating additional profile information when taking into account three wavelength regions. For example the weighting functions for the 88° LOS indicate the largest sensitivity of the measured slant columns considering variations of the vertical profile at 9 km altitude at 362.5 nm and 437.5 nm. The increasing width of the 88° LOS weighting function at 485.0 nm demonstrates the additional profile information as a result of the wavelength dependant Rayleigh scattering. Figure 2b displays the averaging kernels. The averaging kernel demonstrates how much of a change in the true profile is detected by the retrieval algorithm in the retrieved profiles. For example the 9 km averaging kernel shows that the retrieval algorithm is able to detect close to

100% of the change in the true profile. This is the optimum behavior of an averaging kernel which is shown in altitudes where the measurements have sufficient profile information. The results of this sensitivity study show a similar or even better quality of the retrieved profiles compared to most setups consisting of ten LOS and only one wavelength region (350 or 500 nm) shown in Bruns et al. (2004); Bruns (2004). For example the retrieved profiles of the 10 LOS setup at 350 nm (model 1) in Bruns et al. (2004) have a poorer quality in the lower troposphere than those of the 4-3 setup shown in this work. Another example is the 4 LOS setup at 500 nm shown in Fig. 2a. These retrieved profiles have a much poorer quality in the lower troposphere and a poorer vertical resolution. The conclusion of the retrieved profiles from the 4-3 setup having a good quality is supported by Fig. 3b showing the total retrieval error of the retrieved profiles. A larger total retrieval error indicates a poorer quality of the retrieved profiles.

Figure 5 represents the vertical resolution of the retrieved profiles of the 4-3 setup. The FWHM of the averaging kernels is taken as a measure for vertical resolution as suggested in Rodgers (2000). In the lower troposphere a vertical resolution of 3.0 to 4.5 km is to be expected and 2.0 to 3.5 km near flight altitude.

Two physical effects provide vertical information from the measurements: By using measurements taken simultaneously in different lines of sight, different paths through the atmosphere are probed with varying vertical sensitivity. In particular, the measurements pointing close to the horizon have a long light path near the altitude of the aircraft and therefore are very sensitive to absorptions at this height. The second source of profile information is the wavelength dependence of the signal. As result of increased Rayleigh scattering in the UV, the sensitivity to layers close to the surface

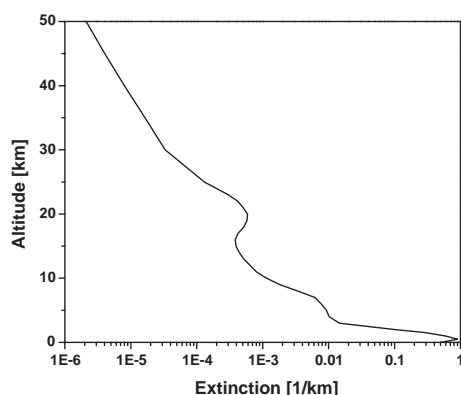


Fig. 4. The vertical extinction profile of the urban aerosol setting.

is reduced compared to measurements at visible wavelengths. By combining retrievals at different wavelengths, some vertical resolution can be obtained even for one viewing direction (see Wang et al., 2004).

5 Results and discussion

In this section the retrieved profiles from AMAXDOAS data will be discussed using meteorological and geographical data. This discussion also includes the comparison of vertical columns calculated from profiles retrieved by APROVAL and vertical columns retrieved from SCIAMACHY data. This comparison was done by Heue et al. (2005) for the same flight. The SCIAMACHY tropospheric vertical columns used in Heue et al. (2005) were calculated by the Excess-method (Richter and Burrows, 2002) where the slant columns retrieved from SCIAMACHY data over a clean air region (Pacific Ocean) at the same latitude are subtracted from the slant columns retrieved over polluted areas to yield tropospheric slant columns. Division by a tropospheric air mass factor yields the tropospheric vertical columns.

Figure 6 presents the retrieved profiles of a part of the flight from 19 February 2003 between 8.350 UTC and 9.305 UTC (see black dots in Fig. 1). During this time 104 profiles have been retrieved with an integration time of 30 seconds each (this translates to an AMAXDOAS footprint of 6.6 km·0.1 km). The upper panel (plot a) shows all profiles retrieved from the measurements taken during this period of time as a contour plot where the x-axis represents the time of the day in UTC and the y-axis represents the altitude in km. The color code indicates the NO₂ VMR in ppbv. The lower panel (plots b through e) shows four examples of retrieved profiles taken from interesting parts of the flight (see colored marks in plot a) where the x-axis represents the NO₂ VMR in ppbv and the y-axis again indicates the altitude in km.

Figure 6a shows tropospheric NO₂ on several occasions. Five tropospheric NO₂ will be discussed in detail here. The

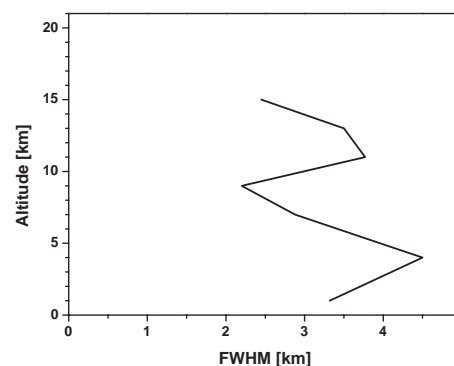


Fig. 5. The vertical resolution (FWHM of the averaging kernels) of profiles retrieved from data using the 4-3 setup.

first observation of tropospheric NO₂ is north of the Alps (see Fig. 7a). The footprint of the first measurement (8.350 UTC) is covering part of the Rhine valley containing two major highways – the Swiss N13 and the Austrian A14. The daily averaged volume of traffic on the Swiss highway N13 is 23 625 (ASTRA, 2003) vehicles for February 2003 on weekdays (19 February 2003 is a Wednesday). The time of crossing the Rhine valley and the Swiss highway N13 was 8.350 UTC (9:21:00 local time) shortly after the morning rush hour. Figure 6b shows the plot of the measurement at 8.350 UTC. It can be seen that the tropospheric NO₂ value of 2.9 ppbv at 8.350 UTC coincides with crossing the Rhine valley and the Swiss highway N13 (see Fig. 7a). Pundt et al. (2005) have shown that major highways are a large source for NO₂ emissions. Figure 8 shows the temperature soundings (Oolman, 2005) of five stations covering a large area including Munich (Germany), Payerne (Switzerland), Milano, Udine, and San Pietro (all Italy). The last three locations do cover the Po valley quite perfectly since Milano is situated on the north western rim, Udine is situated on the north eastern rim and San Pietro on the southern rim of the Po valley only 30 km north east of Bologna. This plot indicates that large parts of Europe and especially the Po valley were subject to a stationary temperature inversion caused by the very stable high pressure system “Helga” situated over southern Scandinavia. This weather situation explains the accumulation of the enhanced NO₂ values observed in the Rhine valley due to the morning rush hour. Since the atmosphere during those weather conditions is very calm, transport will only play a minor role.

The second tropospheric NO₂ event is observed at 8.660 UTC crossing the valley hosting the major transit route for crossing the Alps – the Italian highway A22 (see Fig. 7b). Figure 6c presents the corresponding profile revealing an enhanced NO₂ value of 2.8 ppbv. It has to be mentioned that the next measurement which is crossing the highway A22 directly shows a similar value of enhanced NO₂ (2.3 ppbv).

The third observation (8.725 UTC) of tropospheric NO₂

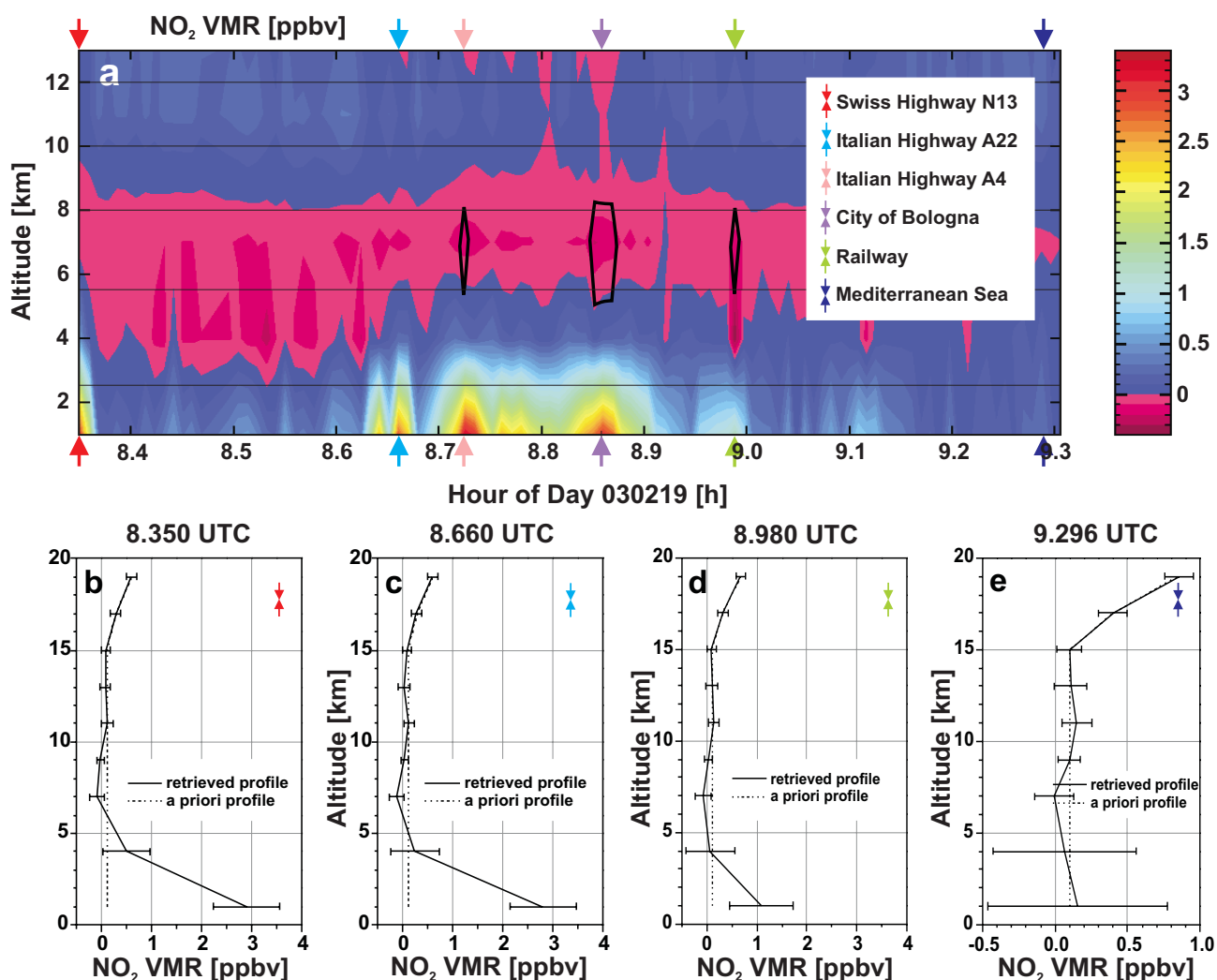


Fig. 6. Retrieved profiles of flight 030219. Plot (a) represents a contour of all retrieved profiles of flight 030219. The thin vertical lines represent the boundaries of the individual layers to be retrieved. The thick lined solid polygons indicate areas of the profile where the profile values added to the error bars are still negative. In other words these profile values are negative even when the error bar is added. Plot (b) shows the retrieved profile of tropospheric NO₂ at 8.350 UTC, plot (c) depicts the retrieved profile of NO₂ in the boundary layer region at 8.660 UTC, plot (d) indicates the retrieved profile of tropospheric NO₂ at 8.980 UTC, and plot (e) shows the retrieved profile of NO₂ in the UTLS region at 9.296 UTC. The dark blue and green arrows mark the positions of the profiles shown in plots (b) and (c) and the red and light blue arrows mark the positions of the profiles represented by plots (d) and (e).

is just south of the Alps near the city of Verona (Italy, population: 260 000; Wikipedia, 2005). The footprint of the measurement (not shown here) is again very close to an Italian highway (A4). It was not possible to find any statistical data on traffic for highway A4. Figure 6a shows the retrieved profile for the measurement at 8.725 UTC with 3.2 ppbv of tropospheric NO₂. The prior measurement shows a similar enhanced tropospheric NO₂ value of 2.3 ppbv. Converting the profile measured at 8.725 UTC into a tropospheric NO₂ vertical column (0–2.5 km) results in a vertical column density of $2.2 \cdot 10^{16}$ molecules/cm². Heue et al. (2005) present tropospheric NO₂ vertical columns measured by the SCIAMACHY instrument on 19 February 2003.

The temporal coincidence of both measurements is very good (within a few minutes). The results of the SCIAMACHY data analysis identified a tropospheric NO₂ vertical column of 2.8×10^{16} molecules/cm² at the geolocation of the measured profile shown in Fig. 6d. Compared to the AMAXDOAS value it is 27% larger which is not very unusual since the footprints of the SCIAMACHY measurements (60 km × 30 km) are much larger than the footprints of the AMAXDOAS measurements (6.6 km × 0.1 km). The larger footprint of the SCIAMACHY measurements result in a higher degree of averaging over a large area. On the other hand the AMAXDOAS measurements show a large variability in boundary layer NO₂ values even in the highly polluted

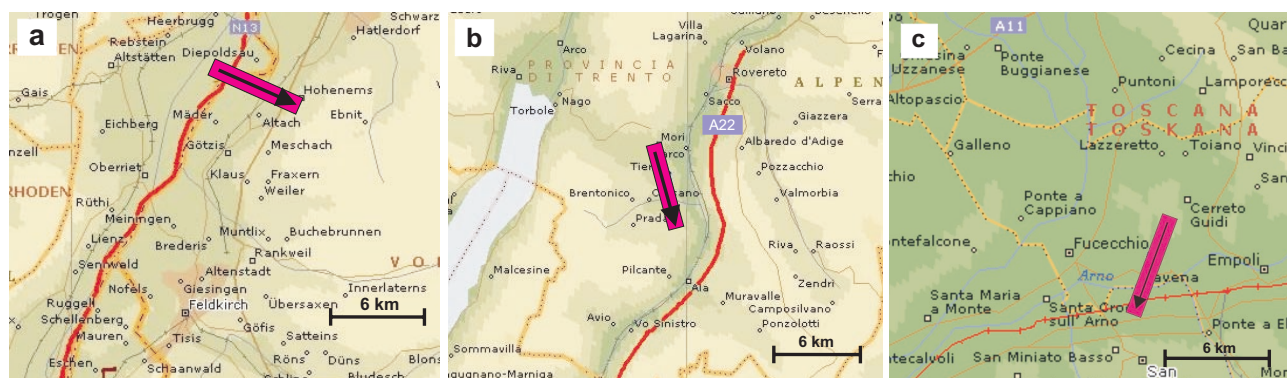


Fig. 7. Maps demonstrating the footprints of the AMAXDOAS measurements shown in Fig. 6. Map (a) depicts the footprint of the measurement taken at 8.350 UTC. This footprint crosses the Swiss major highway N13 (marked in red). Map (b) is a schematic of the footprint measured at 8.660 UTC crossing the Italian major highway A22 (marked in red) which is a major route for crossing the Alps. Map (c) is a schematic of the footprint measured at 8.980 UTC crossing a railway track in Tuscany. These maps have been created using Microsoft Encarta 2001.

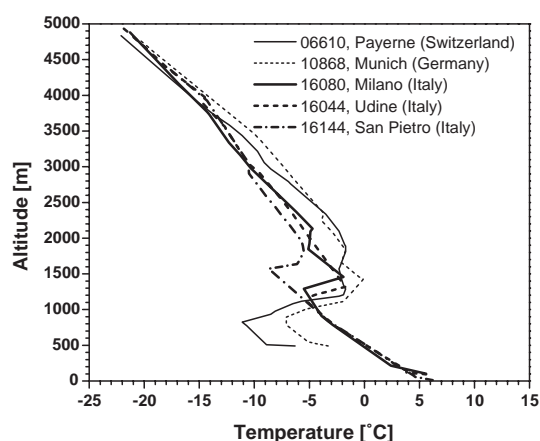


Fig. 8. Stationary temperature inversion caused by the high pressure system “Helga” which was present over large parts of central Europe on 19 February 2003, 12:00 UTC. The data is taken from the website of the University of Wyoming, Department of atmospheric sciences (Oolman, 2005).

Po valley. This implies that there are much higher values of tropospheric NO₂ outside the AMAXDOAS flight track for example the city of Verona which is included in this particular SCIAMACHY footprint.

The fourth event of tropospheric NO₂ occurs over the city of Bologna (8.860 UTC) at the southern rim of the Po valley. Figure 6a shows an enhanced tropospheric NO₂ value of 3.0 ppbv. Large parts of the footprint are observing Bologna city area. Bologna is a city with a population of 370 000 (Wikipedia, 2005). The following measurement has an enhanced tropospheric NO₂ value of almost the same size (2.7 ppbv) since part of this footprint is also covering Bologna city area.

The last event of tropospheric enhanced NO₂ values

shown in Fig. 6a occurs at 8.890 UTC. Figure 6d contains the corresponding profile indicating an enhanced tropospheric NO₂ value of 1.1 ppbv in the boundary layer. Figure 7c presents the location of the footprint of this measurement. It can be seen that it crosses a railway track in Tuscany west of the city of Florence.

Figure 6e shows a profile measured over the clean air area of Mediterranean Sea (9.296 UTC). This profile shows no enhanced NO₂ and was included to demonstrate the response of the profile retrieval to clean air situations. It can be seen that the retrieval algorithm retrieves the a priori information in the troposphere as expected except at 7 km altitude where the retrieved NO₂ value is close to zero. Figure 6a reveals a systematic behaviour regarding too low NO₂ values at this specific altitude but these values are not significantly too low since the upper error boundary is larger than zero for most of the presented profiles. This can be improved by using more LOS in combination with different wavelength regions.

6 Conclusions

The AMAXDOAS instrument is the first to be able to measure tropospheric NO₂ profiles using a remote sensing technique from an airborne platform. This investigation and Bruns et al. (2004) have shown that the combination of four lines of sight and three different wavelength regions is a very good setup to retrieve NO₂ profile information from airborne multi-axis UV/visible scattered skylight measurements using the AMAXDOAS Profile Retrieval Algorithm (APROVAL). This has been demonstrated by a theoretical study focussing on the combination of four lines of sight and three wavelength regions (4–3 setup). The retrieved profiles have a vertical resolution of 3.0 to 4.5 km in the lower troposphere and 2.0 to 3.5 km near flight altitude. The profile information above 13 km altitude is sparse.

In this study the retrieved profiles of a part of the AMAXDOAS flight on 19 February 2003 have been analyzed. The flight took off in Basel (Switzerland) and headed for Tozeur (Tunisia) crossing the Po-valley in Italy which is notorious for significant anthropogenic pollution. Boundary layer values for NO₂ of up to 3.2 ppbv have been detected in this area and were compared to SCIAMACHY tropospheric NO₂ vertical columns from the same day. The comparison of both vertical columns is quite good considering the differences in the size of the footprints of the AMAXDOAS and SCIAMACHY measurements. All major events of tropospheric NO₂ shown in Fig. 6 could be assigned to anthropogenic sources. The weather situation of a stationary temperature inversion over large parts of Europe especially the Po valley is a very reasonable cause for the enhanced tropospheric NO₂ values found nearby the respective footprints of the AMAXDOAS measurements since transport is very unlikely during such a stable weather condition.

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