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Airborne multi-axis DOAS measurements of tropospheric SO_2 plumes in the Po-valley, Italy

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Abstract

During the second FORMAT (FORMaldehyde as A Tracer of oxidation in the troposphere) campaign in 2003 the airborne multi-axis DOAS instrument (AMAXDOAS) performed spectroscopic measurements of SO_2 from the city of Mantova and the power plant Porto Tolle using scattered sun-light during two flights on 26 and 27 September 2003. Measurements were performed in 10 different viewing directions, providing information on the vertical SO_2 distribution and the SO_2 vertical column. The SO_2 emission flux from the power plant Porto Tolle was calculated to 1.6×10^{25} molec cm^{-2} (1.7 kg s^{-1}) and was found to be the same on both measurement days, and also comparable to official emission data, which quote 2.25×10^{25} molec s^{-1} (26 September) and 2.07×10^{25} molec s^{-1} (27 September). Over the city of Mantova, the observed SO_2 vertical columns were 1.1×10^{16} molec cm^{-2} and 1.9×10^{16} molec cm^{-2} on 26 and 27 September, respectively. This is in good agreement with ground-based measurements of 5.9 ppbv and 10.0 ppbv which correspond to 1.2×10^{16} molec cm^{-2} and 2.2×10^{16} molec cm^{-2}.

1. Introduction

Sulphur Dioxide, SO_2 is directly emitted into the atmosphere by volcanoes and also produced by the oxidation of sulphur containing gases in the atmosphere. The main anthropogenic sources of SO_2 are combustion of fossil fuels, for example in electric power plants, refinery emissions, and to a lesser degree biomass burning. In the absence of clouds, SO_2 is converted to H_2SO_4 through homogeneous gas-phase reactions initiated by the hydroxyl radical. Generally only a minority of SO_2 is oxidized in air, the rest is removed by dry deposition. In the presence of clouds a fraction of the SO_2 is dissolved into cloud droplets and oxidized to sulphate ions, SO_4^{2-}, by trace amounts of oxidizing agents such as hydrogen peroxide, H_2O_2, and O_3 that are present in the airborne droplets. Eventually it is removed by wet deposition (Wayne, 1991).

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SO_2 itself is a respiratory irritant, the effect appearing at concentrations above 1 ppm (Wayne, 1991). SO_2 is oxidized to H_2SO_4 and contributes to acid rain. It increases acidity in the aquatic ecosystem and is harmful for soil and vegetation. SO_2 inhibits photosynthesis in plants and reduces plant growth.

SO_2 concentrations are routinely measured by air quality monitoring networks, such as ARPAV and Lombardia air quality network (http://www.arpa.veneto.it/indice.htm; http://www.ambiente.regione.lombardia.it/webqa/QualitAmbiente.htm). In-situ measurements of SO_2 were part of many air-borne campaign measurements (e.g. Tscherwenka et al., 1998; Svensson and Klemm, 1998; Thornton et al., 2002; Tu et al., 2003). Remote sensing measurements of SO_2 from space have been performed using TOMS (Krueger et al., 1995; Carn et al., 2004), GOME (Eisinger and Burrows, 1998; Khokhar et al., 2005[1]), SCIAMACHY (Afe et al., 2004) and AIRS (Carn et al., 2004) measurements, but are mainly restricted to volcanic eruptions or large scale pollution. The COSPEC (COrrelation SPECtrometer) technique developed in the late 1960s has also been used to study total emissions of SO_2 and NO_2 from various sources, e.g. industrial emissions (Millan et al., 1969; Hoff and Millan, 1981) and volcanic plumes (Hoff, 1992).

The airborne multi-axis DOAS instrument (AMAXDOAS) has previously been used on board the high flying aircraft DLR-Falcon in campaigns dedicated to the validation of the SCIAMACHY instrument on ENVISAT (Bovensmann et al., 1999). Several papers have demonstrated its use for measurements of the tropospheric NO_2, and the validation of SCIAMACHY NO_2 (Heue et al., 2004; Wang et al., 2005; Fix et al., 2004). The possibility to retrieve vertical profiles from the measurements has also been reported (Bruns et al., 2004). In summer 2002, the AMAXDOAS instrument was for the first time operated onboard the low flying aircraft Partenavia, to measure HCHO and

[1]Khokhar, M. F., Frankenberg, C., Van Roozendael, M., Beirle, S., Khl, S., Richter, A., Platt, U., and Wagner, T.: Satellite observation of atmospheric SO_2 from volcanic eruptions during the time period of 1996 to 2002, Adv. Space Res., submitted, 2005.

NO_2 abundances in urban plumes (Pundt et al., 2005a)[2]. During the second FORMAT campaign, the viewing directions were optimized for the measurements of plumes from point sources (Pundt et al., 2005b)[3].

In this study we focus on measurements of the SO_2 flux from the power plant Porto Tolle and the SO_2 concentration at the city of Mantova. The measurements were performed using the AMAXDOAS instrument onboard the aircraft Partenavia in the Po-valley, northern Italy in September 2003. For the power plant plume, the SO_2 emission rate was derived and compared with official emission data. For the city of Mantova, the SO_2 vertical columns were determined and compared with ground-based in situ measurements.

2. AMAXDOAS setup during the second FORMAT campaign

The AMAXDOAS instrument consists of two grating spectrometers, one operating in the UV between 300–440 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 outside of the aircraft, one on the top and one on the bottom (Wagner et al., 2001). Measurements are performed in ten viewing directions, where the zenith direction is denoted as 180°, the nadir direction is 0°, and the flight direction 90°. The viewing directions are shown in Fig. 1. The signals from the ten directions are detected simultaneously with CCD imaging detectors. During measurements the CCD detectors are cooled down to −30°C and the spectrometers are thermally stabilized at about 40°C to prevent wavelength drifts during the flight. The UV spectra images were

[2]Pundt, I., Heue, K.-P., Wang, P., Richter, A., Friedeburg, C. V., Bruns, M., Laepple, T., Wagner, T., Burrows, J. P., and Platt, U.: Airborne Multi-Axis-DOAS measurements of formaldehyde of the photochemical plume of Milan city, paper in preparation, 2005.

[3]Pundt, I., Heue, K.-P., Song, B.-C., Richter, A., Wang, P., Bruns, M., Platt, U., Burrows, J. P., and Wagner, T.: Airborne Tomographic Measurements of NO_2 Plumes from Point sources using the AMAX DOAS instrument, paper in preparation, 2005.

recorded with 10 s integration time, and dark current and line lamp calibration measurements were performed after the flight. In order to improve the signal to noise ratio, the measured spectra were averaged over 1 min intervals before further analysis.

3. Data analysis

3.1. SO_2 slant column

The data analysis is based on the Differential Optical Absorption Spectroscopy (DOAS) method (Platt, 1994). For the SO_2 fit, the spectral window of 316.5–325.5 nm was selected. Two O_3 cross sections at 293 K and 221 K, respectively (Burrows et al., 1999), an NO_2 cross section at 293 K (Burrows et al., 1998), the SO_2 cross section at 295 K (Vandaele et al., 1994), the HCHO cross section (Meller and Moortgat, 2000) and a ring spectrum (Vountas et al., 1998) were included in the fit. For each measurement direction, a background spectrum taken in the same viewing direction during the same flight was used. The background spectra were chosen to be close to the SO_2 plume to minimise the effect of instrumental changes but in a region where low SO_2 is expected. The result of the DOAS analysis is the differential slant column, which is the slant column relative to the background spectrum. With our background spectrum criteria, the differential slant column is actually the SO_2 slant column of the plume.

3.2. Airmass factor calculation

The slant columns retrieved with the DOAS method have to be converted to vertical columns, usually by dividing through appropriate air mass factors (AMF). The AMF is defined as the ratio of the slant column and the vertical column of the absorber. In this study, AMFs were calculated with the radiative transfer model SCIATRAN 2.0 full spherical version (Rozanov et al., 2001) for all viewing directions at flight altitude (600 m). The viewing angles were corrected for the pitch and roll of the aircraft. However, for both the zenith and nadir viewing directions the effect of this correction is small.

As the power plant Porto Tolle is located near the coast, a maritime aerosol is expected with the exception of the exhaust plume itself which has much higher aerosol loading. The aerosol type near the city Mantova was assumed to be urban. The aerosol optical depth used in the radiative transfer model is about 0.35 (at 550 nm) which is similar to the aerosol optical thickness given by MODIS data on that day (Kaufman and Tanre, 1998). The results were tested with O_4 slant columns and good agreement between different viewing directions was found with these settings. Details on the method used for determination of the aerosol settings can be found in (Heckel et al., 2004) and (Wang et al., 2005). Throughout the measurements, the sky was cloud free, and therefore no clouds were included in the radiative transfer calculations. The surface albedo was set to 0.02 at 320 nm, the central wavelength of the SO_2 fitting window. Using these settings, air mass factors were calculated for solar zenith angles between 40° to 75° with 5° intervals at 320 nm.

During the measurements, two kinds of emission plumes of SO_2 were sampled, one from a power plant, and one from a city. Therefore, two sets of AMFs were calculated with different types of SO_2 profiles. To calculate the SO_2 AMFs for the city, the profile was assumed to be well mixed between 0 and 500 m. This choice is based on the fact that no significant SO_2 slant column increase was observed above flight altitude as discussed in Sect. 4.1. The AMFs for the plume near the power plant were calculated with a well mixed profile below 1.7 km, as the boundary layer height was between 1.5 and 1.8 km based on the dew point profile and other trace gases profiles measured onboard another aircraft nearby, the Ultralight (Junkermann, 2005) on 26 September during the comparison flight.

3.3. SO_2 flux calculation

To calculate the emission flux from the power plant Porto Tolle, a simple formula based on the integrated amount of SO_2 in the transect measured from the aircraft and the

wind speed perpendicular to the flight direction can be used:

$$Flux = v_{\text{aircraft}}\, v_{\text{wind}}\, \cos\theta \int_{t_1}^{t_2} VC(t)\, dt\,, \tag{1}$$

where v_{aircraft} is the velocity of the aircraft, v_{wind} is the wind speed, θ is the angle between the wind direction and the flight direction, VC is the SO_2 vertical column and $t_1..t_2$ is the time interval flown in the plume (White et al., 1976; Trainer et al., 1995; Melamed et al., 2003). Since there were no clouds and humidity was low (relative humidity about 60%), the SO_2 emitted from the power plant was probably not converted to H_2SO_4 or removed by deposition very fast. If both the wind direction and wind speed are constant throughout the boundary layer, the measurement should give a good approximation of the emission flux from the power plant.

4. Results and discussion

4.1. Enhanced SO_2 slant columns at Porto Tolle and Mantova

On 26 and 27 September 2003 the flight started from Milan to the south, lead over Pavia, turned to the east, over Cremona and Mantova, then turned around the power plant Porto Tolle, and back to Milan. To measure the plume from the power plant, the aircraft flew around the stacks with a roughly 3 km radius as illustrated in Fig. 2. The flight routes taken on the two days around the power plant Porto Tolle are almost the same. On 26 September on the way back from Porto Tolle the aircraft also flew around another power plant at Sermide and Ostiglia (close to 45.0° N, 11.2° E), and did a comparison flight with other aircraft at 45.0–45.2° N, 11.45° E. Measurements with the Ultralight aircraft performed during the comparison flight indicate that NO_2 and HCHO were roughly well mixed below 1.5–1.8 km. The flight altitude was mainly about 600 m except for the intercomparison flight where it was at about 1.8 km. The flight started at 09:00 UT on 26 September, and at 08:55 UT on 27 September.

Enhanced SO_2 values were clearly identified both at Porto Tolle and Mantova. An example of the DOAS fit is shown in Fig. 3. That measurement was in zenith viewing direction, near the power plant Porto Tolle at 10:24 UT on 27 September, at a solar zenith angle of 47.26°. The background spectrum used was measured at 44.94° N, 11.36° E, which is about 100 km west of Porto Tolle. The error of the fit was about 12%. The background spectrum for 26 September was measured at 45.04° N, 12.40° E, the upwind direction of Porto Tolle power plant. For the analysis the assumption is made, that the background spectra contain no SO_2 absorption signature.

The SO_2 slant columns were measured in 10 viewing directions. Three representative viewing directions are shown in Fig. 4. On 26 September, three SO_2 plumes were measured at about 9.6 (09:36) UT, 9.9 (09:54) UT, 10.6 (10:36) UT near the city Cremona, Mantova, and the power plant Porto Tolle, respectively. On 27 September the same plumes were measured at about 9.3 (09:18) UT, 9.6 (09:36) UT and 10.4 (10:24) UT. The SO_2 plume from the power plant was observed both in the upward and downward viewing directions. In contrast, the plumes of the cities of Cremona and Mantova were only measured in the downward viewing directions. At Porto Tolle, the SO_2 slant columns of the zenith viewing direction were similar on the two days, which indicates that the emissions from the power plant did not change very much as the wind speed was also similar. The SO_2 slant columns in the 97° and 83° viewing direction were larger than that in the zenith viewing direction due to the enhanced optical path through the plume. Over Mantova the SO_2 slant columns measured on 27 September are about two times that of 26 September. The SO_2 plume over Cremona is smaller than the other two plumes. From the SO_2 time series it is also obvious that the slant columns have relatively large variations in the background, which makes it difficult to detect small SO_2 plumes. On the way back from Porto Tolle, on 26 September the flight didn't cross Cremona, and on 27 September the flight did not cover Mantova.

The NO_2 slant columns in the three viewing directions on 26 September are shown in Fig. 5 for comparison as power plant emissions of SO_2 and NO_2 should be well correlated and NO_2 measurements have much higher accuracies. The fitting window

selected for the NO_2 retrieval is 345–380 nm, which is completely independent from the SO_2 fit. Several different NO_2 emissions contribute to the NO_2 signal and it is difficult to identify individual sources. However, the NO_2 plume from the power plant Porto Tolle can clearly be identified at about 10.6 (10:36) UT. As in the case of SO_2, the NO_2 plume from the power plant is also measured in all viewing directions. The 97° viewing direction has a similar amount of NO_2 as the 83° viewing direction. The similarity of the NO_2 and SO_2 measurements adds confidence to the SO_2 measurement.

4.2. SO_2 emission flux at power plant Porto Tolle

The AMAXDOAS measurements show enhanced SO_2 close to the Porto Tolle power plant. To calculate the emission flux from these measurements, it has to be assured that the plume from the power plant was fully sampled, and the measured slant columns have to be converted to vertical columns to derive the total amount of SO_2 in the plume transect. For the conversion of slant to vertical columns, an assumption has to be made on the vertical distribution of the SO_2. For this, both the AMAXDOAS measurements themselves and in-situ surface measurements were used.

In Fig. 6 SO_2 slant columns (97° viewing direction) are shown around the power plant along the flight track. As can be seen, the location of the SO_2 plume was to the south of the power plant as expected from the wind direction.

On 26 September the wind speed at 10:28 UT (at 44.92° N, 12.28° E) was about 4.5(±2) m/s and the wind direction 345°(±30°) (north is 0°). On 27 September the wind speed at 10:28 UT (at 44.92° N, 12.11° E) was also about 4.5(±2) m/s, however, the direction was 18°(±30°). Both wind speed and direction were measured on the aircraft during flight. The position of the observed plume is in good agreement with the wind direction on 26 September. The plume was displaced to the west relative to the wind direction on 27 September, indicating a large uncertainty in the wind direction measured on the aircraft. The similar weather conditions are one reason for the similarity of the slant columns measured on the two days in the zenith viewing direction. As the slant columns measured in the 97° viewing direction were also similar on the two

days, it can be assumed that the vertical position of the plume was also comparable.

The Electric Energy Board (ENEL) operates one central meteorology station at the center of the Porto Tolle power plant and eight air quality measurement stations around the power plant, the one closest to our flight track being at Scardovari (44.9° N, 12.46° E), at the south of the power plant, see Fig. 6. As shown in Fig. 7, the SO_2 concentration at Scardovari showed a large peak between 10:00 and 15:00 UT on 26 September. The station is located downwind of the power plant (the surface wind direction was from north to south measured at the central meteorological station), almost at the centre of the plume, according to the AMAXDOAS measurements. The very large concentrations observed at the station during the AMAXDOAS overpass show that the plume reached the surface and indicate that the boundary layer was probably well mixed. On 27 September, the SO_2 concentration at Scardovari did not vary significantly during the day, also in agreement with the AMAXDOAS measurements that show no indication for enhanced SO_2 close to the station. On 27 September, the Scardovari station did not sample the plume because the surface wind direction was from west to east before 10:00 UT and then turned to east to west, the wind speed being low throughout the day.

The SO_2 of the power plant Porto Tolle is emitted from a stack of 250 m altitude at a temperature of about 130°C (http://www.gruppoverdier.it/documenti.php). Thus, the plume can easily be transported to higher altitude and in fact could be measured in all AMAXDOAS viewing directions. At noon the turbulence in the boundary layer is usually strong, and the SO_2 could be well mixed in the boundary layer a few kilometres downwind of the stack. The measurements of the SO_2 peak were made at distances of about 5 km (26 September) and 11 km (27 September) from the stack, and therefore the SO_2 profile was assumed to be well-mixed below 1.7 km, the top of the boundary layer.

The SO_2 vertical columns for the power plant plume are calculated from the zenith viewing direction, as this measurement has a high signal to noise ratio and is insensitive to the relative azimuth angle of the sun. The SO_2 AMF in zenith viewing direction is

about 1.1 for the power plume using the assumptions described above. Thus, the SO_2 vertical column near the power plant is 4×10^{16} molec cm^{-2}. The SO_2 vertical columns measured on both days near the Porto Tolle power plant are shown in Fig. 8. For comparison, the SO_2 vertical columns calculated from the measurements in nadir viewing direction are also shown. The SO_2 AMF for the plume in the nadir viewing direction is about 2.4.

The SO_2 vertical columns calculated from zenith and nadir directions are very similar for both measurements, although the data in the nadir direction are noisy. Therefore, only the zenith vertical columns were used to calculate the emission flux. The measurements in the other viewing directions have also been evaluated yielding maximum vertical columns in the range of 2-4$\times10^{16}$ molec cm^{-2}.

The AMAXDOAS SO_2 vertical columns are in good agreement with the ENEL in situ measurements at Scardovari. The in situ SO_2 concentration at 10:50 UT was about 27 μg m^{-3} or 10.3 ppbv (parts per billion volume mixing ratio) on 26 September. Converting the mixing ratio to a vertical column with the assumption of a well-mixed profile from the surface up to 1.7 km yields 4.4×10^{16} molec cm^{-2}. The same calculation for our measurements yields a mixing ratio of 9.4 ppbv at ground. This value represents the mixing ratios coming only from the power plant, because the AMAXDOAS measurement is taken relative to a reference on the upward side of the plume. If we add an SO_2 background level of about 1 ppbv (see Fig. 7: The background concentration is about 2.7 μg m^{-3} which corresponds to 1 ppbv.), our measurements quote 10.4 ppbv at ground. This is in excellent agreement with the 10.3 ppbv given by the ENEL station.

Using formula (1), the SO_2 emission flux from the Porto Tolle power plant can be calculated from the measurements. The half width of the spatial extent of the plume (the half with in time multiplied with the aircraft velocity) is multiplied with the peak column density, thus obtaining the integrated SO_2 contents per unit length of plume (in e.g. molecules cm^{-1}). Multiplying this quantity with the wind velocity yields the flux in molec s^{-1} or kg s^{-1}. The time needed to cross the plume was about 6 minutes at a flight speed of 230 km h^{-1}. With a wind speed of 4.5 m s^{-1}, the SO_2 flux

is 1.57×10^{25} molec s^{-1} on 26 September and 1.6×10^{25} molec s^{-1} on 27 September. Thus, the AMAXDOAS determined SO_2 fluxes are consistent for the two days.

Hourly averaged SO_2 emission data are also measured by the power plant Porto Tolle. The SO_2 concentration of the power unit groups 1-2-3 was about 1500 mg Nm^{-3} at 11 UT on both 26 and 27 September. As there were no measurements for unit 4 on 26 and 27 September, the averaged concentration between 11 September and 16 November was used, which was 317 mg Nm^{-3}. The SO_2 concentration and power output were very stable with variations being smaller than 1% during the day. The gas flow for each of the power units is calculated from the output power, the burned fuel and its efficiency. The SO_2 emission flux is calculated from the gas flow and the SO_2 concentration. The resulting SO_2 emission flux is 2.25×10^{25} molec s^{-1} on 26 September and 2.07×10^{25} molec s^{-1} on 27 September.

Uncertainties in the SO_2 emission flux derived from AMAXDOAS measurements are introduced by the uncertainty of the SO_2 vertical columns, the wind speed, wind direction, aircraft speed and the time needed to cross the plume. In this case study, the error is dominated by the uncertainty of wind speed and direction. The error bars on the wind speed are ±2 m s^{-1} and ±30° for the wind direction, which will lead to total error of about 50%. There are also significant variations of up to 5×10^{15} molec cm^{-2} SO_2 in the background measurements, which is about 12% of the slant column. The SO_2 calculated here is the SO_2 in the plumes relative to the background. If there is any SO_2 in the background, the AMAXDOAS measured SO_2 will be too small. The fit error in the SO_2 slant column was between 15–50% depending on the SO_2 signal. The SO_2 emission flux derived from AMAXDOAS and the power plant measurements are summarized in Table 1.

4.3. SO_2 plumes over the city of Mantova

Enhanced SO_2 over Mantova could only be observed in the downward viewing directions, indicating that the source of the SO_2 is close to the surface. At least, no SO_2

had been transported above 600 m, the flight altitude, according to our detection limit. Judging from the AMAXDOAS weighting functions for the SO_2 measurements in the zenith and 97° viewing directions, the lack of SO_2 signal in the upwards viewing directions indicates that the plume was lower than 500 m. Accordingly, the AMFs were calculated with a profile where SO_2 is located only in the lowest 500 m.

The SO_2 vertical columns measured over the city of Mantova are shown in Fig. 9. The peaks at 9.9 (09:54) UT on 26 September and 9.6 (09:36) UT 27 September are signals from the SO_2 pollution at Mantova. The peak at 9.6 (09:36) UT on 26 September was close to the city of Cremona. The SO_2 vertical columns measured on 27 September are twice as large as those observed on 26 September. The wind directions on 26 and 27 September were similar (south-westerly), but on 26 September the wind speed was much larger than on 27 September before 10:00 UT, see Fig. 10. Low wind velocities usually contribute to the accumulation of SO_2 which probably is the reason or the larger values measured on 27 September.

The closest in-situ station along the flight track at Mantova is the station at 10.82° E, 45.16° N (No. 542). The hourly averaged SO_2 concentration measured at this station between 10:00 and 11:00 UT on 26 September was 5.9 ppbv and 10.0 ppbv on 27 September (http://www.ambiente.regione.lombardia.it/webqa/QualitAmbiente.htm). Assuming that the SO_2 is well mixed below 500 m, this corresponds to vertical columns of 1.09×10^{16} molec cm^{-2} and 1.85×10^{16} molec cm^{-2}, respectively. The AMAXDOAS measurements are 1.2×10^{16} molec cm^{-2} and 2.2×10^{16} molec cm^{-2}, which is in very good agreement with the in situ measurement.

5. Conclusions

In this study, the first airborne multi-axis DOAS measurements of SO_2 pollution are reported. The measurements were performed as part of the second FORMAT campaign in September 2003 in the Po valley, Italy, and covered both a power plant (Porto Tolle) and two cities (Mantova and Cremona).

To interpret airborne remote sensing measurements, information is needed on the vertical position of the absorbing species. The off-axis data of the AMAXDOAS measurements proved to be useful to determine plume altitudes which in turn could be used to establish total columns. At the power plant Porto Tolle, both SO_2 and NO_2 were detected in all viewing directions at a cruising altitude of 600 m, indicating that the plume was well mixed in the boundary layer. This is supported by high concentrations reported from surface in-situ measurements. In contrast, SO_2 enhancement over the city of Mantova was detected in the downwards viewing directions only, so that the plume was assumed to be below 500 m.

Using these mixing heights, SO_2 vertical columns were derived from the AMAXDOAS measurements. The values over Mantova were compared to in-situ measurements, and good agreement was found on both days, highlighting the sensitivity of the measurements.

For the Porte Tolle power plant, the AMAXDOAS data were used to derive an estimate of the power plant emissions. The derived value of $1.60 \pm 0.8 \times 10^{25}$ molec s^{-1} is similar on both measurement days and is comparable to the official emission data of $2.25/2.07 \times 10^{25}$ molec s^{-1}. The advantage of the airborne measurements is, that neither the exact vertical position of the plume, nor the distance of the measurement from the stack need to be known to establish the emissions. Also, the measurement can be performed for any wind direction whereas the surface network will only pick up SO_2 for well mixed plumes passing over the measurement site.

The errors of the estimated emission flux are relatively large, mainly due to uncertainties in wind speed and direction but also as a result of SO_2 measurement errors in particular for smaller values. This could be improved in future measurements by using better meteorological measurements and by optimising the spectrometer for the SO_2 retrieval.

Our measurements and the comparison of the results with independent data demonstrate that the AMAXDOAS instrument is a very useful tool for air quality monitoring in a large number of applications ranging from urban pollution to point sources.

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Table 1. SO_2 emission of the Porte Tolle power plant and surface concentrations inside the plume derived from the AMAXDOAS measurements in comparison with the data measured by the ENEL (* on 27 September the ground station was located outside the emission plume).

Date	AMAXDOAS near Scardovadi ground station (mixing ratio, ppbv)	Insitu Data Scardovadi ground station (mixing ratio, ppbv)	AMAXDOAS Porto Tolle emission rate (10^{25} molec s^{-1})	Insitu Data Porto Tolle emission rate (10^{25} molec s^{-1})
26 September	10.4 (±1.2)	10.3	1.57 ±0.8	2.25
27 September	–	1	1.60 ±0.8	2.07

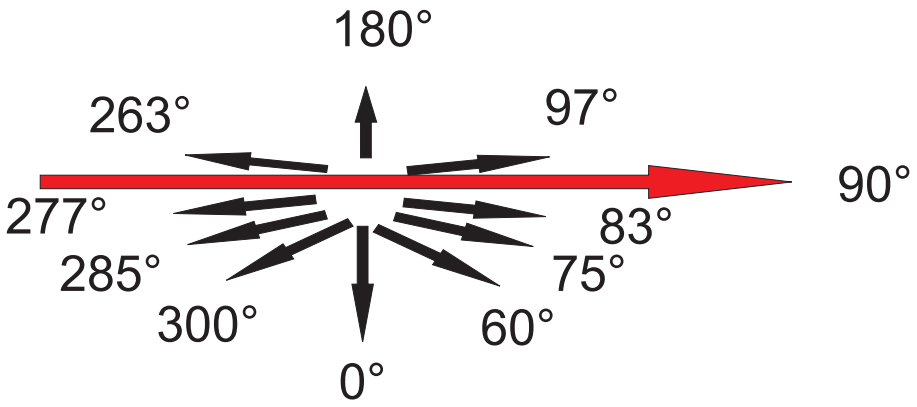

Fig. 1. AMAXDOAS telescope viewing directions. The flight direction is indicated by the red arrow (90°). The ten directions are in the same vertical plane.

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5, 2017–2045, 2005

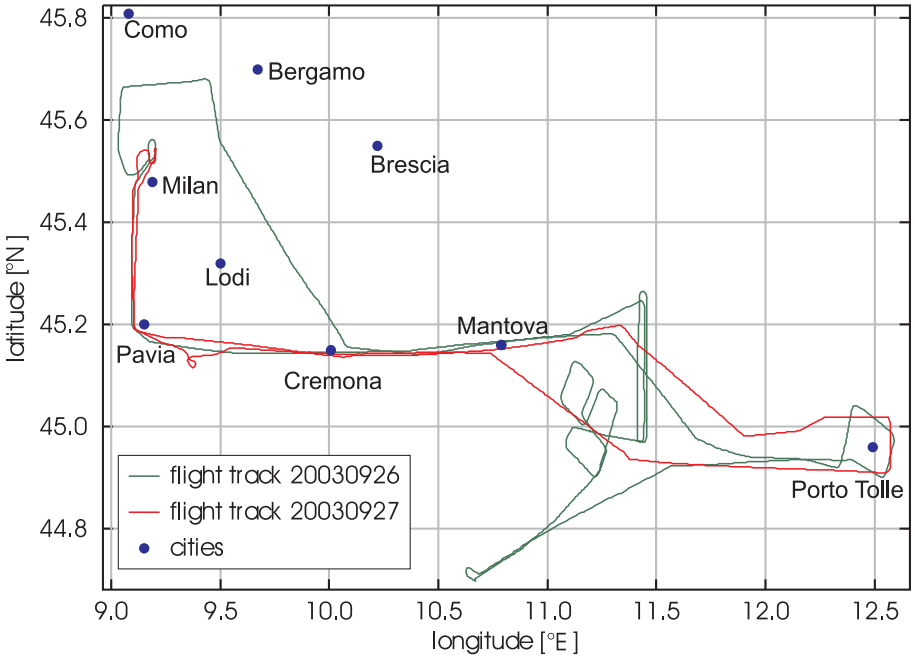

Fig. 2. AMAXDOAS flight tracks on 26 and 27 September 2003.

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5, 2017–2045, 2005

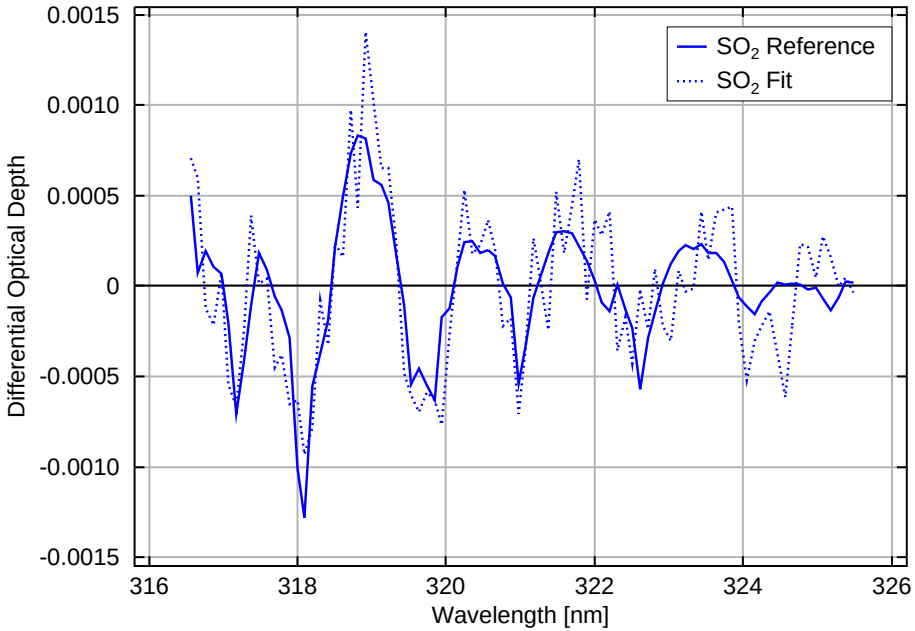

Fig. 3. An example of a SO_2 fit, on measurements close to Porto Tolle at 10:24 UT on 27 September. The solid line is the scaled laboratory reference, the dotted line the result of the fit after subtraction of all other absorbers.

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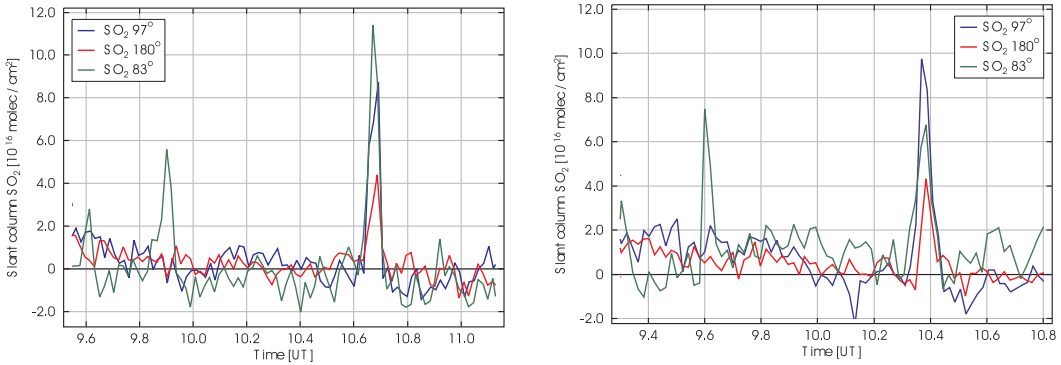

Fig. 4. SO_2 slant columns measured by AMAXDOAS on 26 September 2003 (left) and 27 September 2003 (right) for the flight from Cremona to Mantova, to the Porto Tolle power plant and back to the airport Reggio nell' Emiglia.

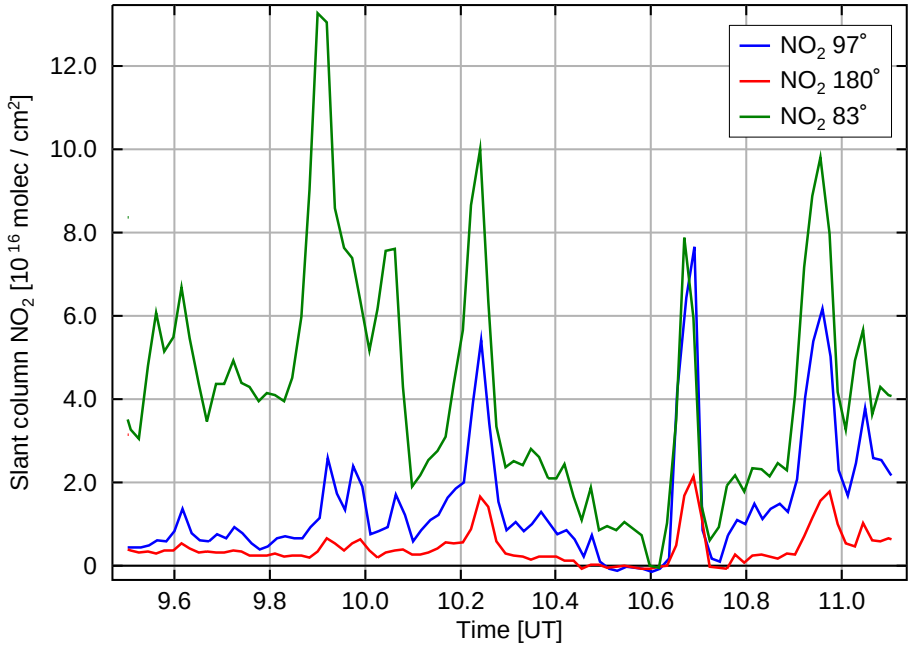

Fig. 5. AMAXDOAS NO_2 slant columns measured on 26 September 2003. The part of the flight shown is from Cremona to Mantova to the Porto Tolle and back to the airport Reggio nell' Emiglia.

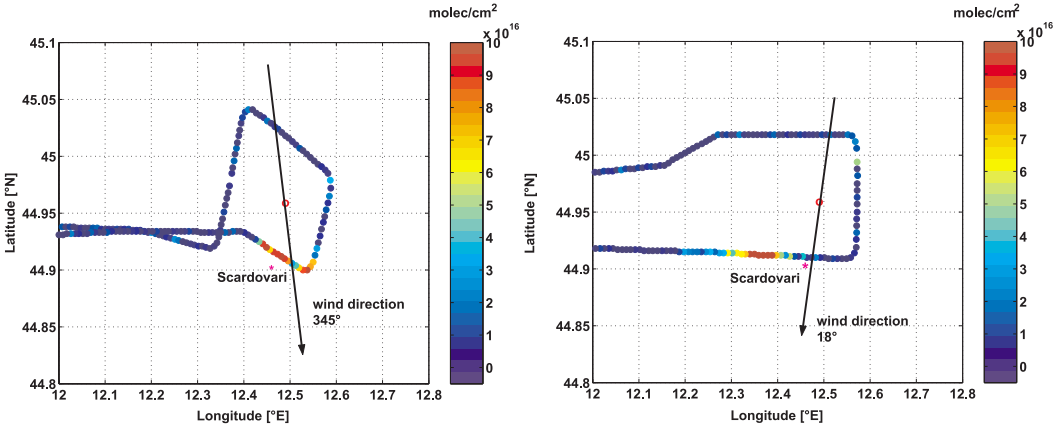

Fig. 6. SO_2 slant columns measured around the Porto Tolle power plant (red circle) in the 97° viewing direction along the flight track on 26 September, (left) and 27 September 2003 (right). Also indicated is the closest ENEL air quality measurement station at Scardovari.

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5, 2017–2045, 2005

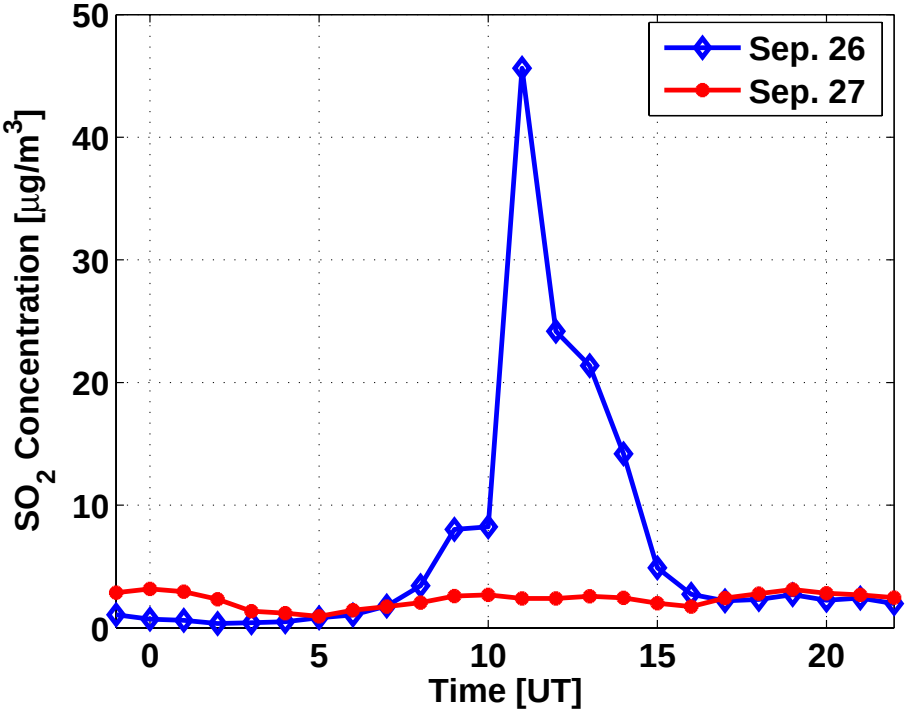

Fig. 7. Hourly averaged SO_2 concentration measured at the ENEL surface station Scardovari on 26 and 27 September, 2003.

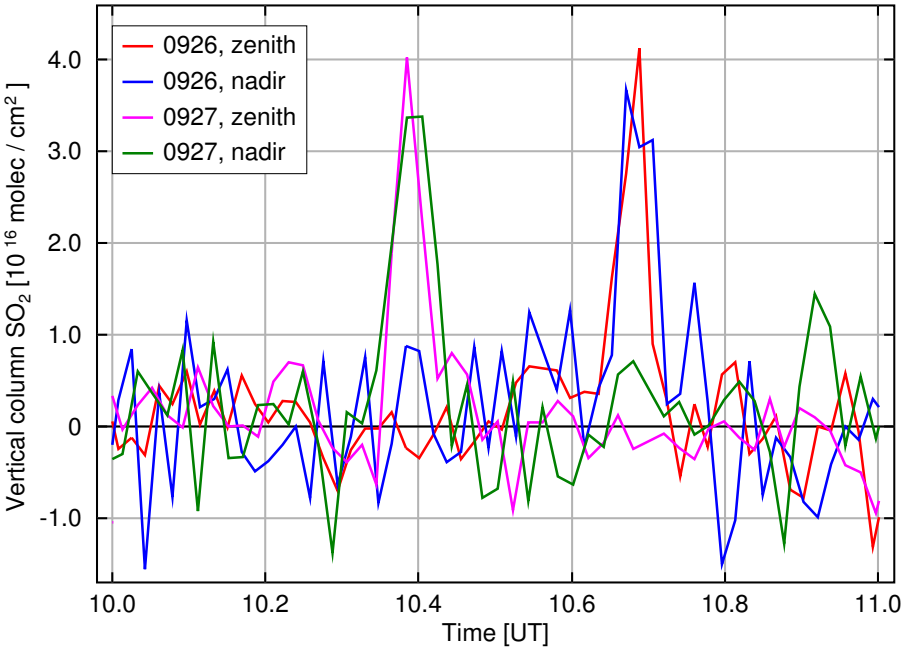

Fig. 8. SO_2 vertical columns measured by AMAXDOAS around the Porto Tolle power plant on 26 and 27 September 2003. Both zenith and nadir measurements are shown.

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5, 2017–2045, 2005

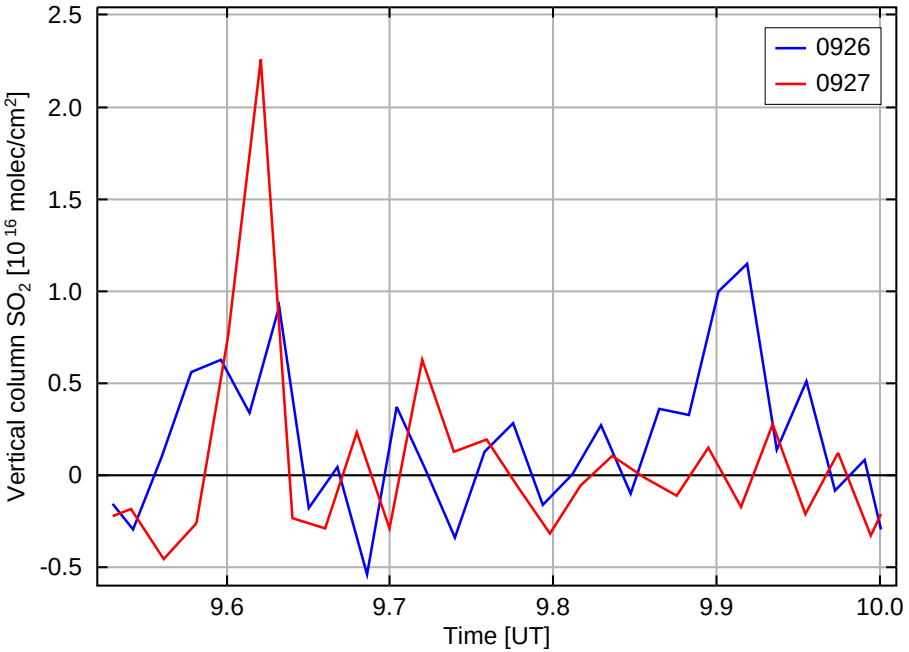

Fig. 9. SO_2 vertical columns measured over Mantova on 26 and 27 September 2003. The peaks at 9.9 UT on 26 September and 9.6 UT 27 September were located over Mantova. The peak at 9.6 UT on 26 September was close to Cremona.

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5, 2017–2045, 2005

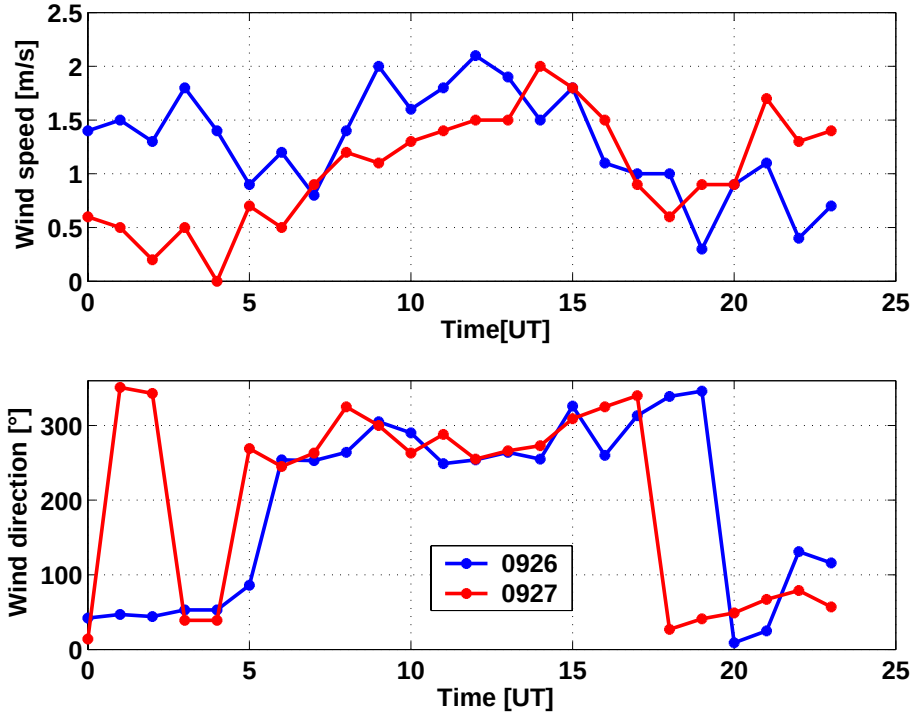

Fig. 10. Wind speed and direction at Mantova (station No. 542) on 26 and 27 September 2003 (data from http://www.ambiente.regione.lombardia.it/webqa/QualitAmbiente.htm).

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5, 2017–2045, 2005