Mapping of tropospheric trace gas concentration distributions from ground and aircraft by DOAS-tomography (Tom-DOAS)

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Abstract – An overview is given on the novel DOAS Tomography technique, its capabilities and the measurements and simulations performed so far. With this technique, column densities of one or several trace gases are measured by DOAS (Differential Optical Absorption Spectroscopy) along multiple (10-400) light paths, and reconstructed to concentration distributions by tomographic inversion. In the framework of the German BMBF project "Tom-DOAS" and the European project "Format", tomographic measurements have been conducted from the ground using Multibeam Longpath DOAS instruments and from aircraft using an AMAX (Airborne Multi AXis) DOAS instrument.

Keywords: DOAS, tomography, atmosphere, NO_2, aircraft, trace gases, mapping

1. INTRODUCTION

Differential optical absorption spectroscopy (DOAS) (e.g. Platt, 1994) is a path-integrating measurement technique for atmospheric trace gases like ozone, nitrogen oxides, SO_2, halogen oxides (BrO, IO, OClO) and many hydrocarbons. For DOAS, a continuous artificial or natural light source (sun, moon, …) is used. Because of the characteristic absorption features it is possible to measure several trace gases simultaneously (e.g. NO_2, SO_2, HCHO, HONO, and ozone in the UV region).

In a typical so-called long-path (LP) setup a beam of white light, emitted by a telescope, travels through the atmosphere to a retro-reflector and back to the telescope. Absorption patterns in the received light allow determining the column density (the integrated concentration) of the trace gases along the light path. LP-DOAS was already used to produce one-dimensional vertical trace gas profiles by placing retroreflectors on a mountain at different heights (Platt, 1978) or on balloons (e.g., Veitel et al., 2002).

A DOAS tomography setup consists of a number of intersecting or non-intersecting light paths, which serve to determine the trace gas distribution in the area of the light paths (Fig. 1).

In contrast to the use of artificial light sources described above, the Airborne Multi AXis (AMAX) DOAS instrument measures passively the sunlight which is scattered in the earth's atmosphere and/or reflected on the ground. During the Tom-DOAS project we use several small telescopes which are directed in various directions and which capture the light simultaneously. Each measurement provides results from several different paths throughout the atmosphere (Fig. 2). As the aircraft moves on from measurement to measurement, data for a vast number of intersecting lightbeams is recorded.

In comparison to other tomographic applications like in medicine, groundbased DOAS-tomography has only a limited number (10-100) of well known light paths. Therefore the reconstruction technique is not time critical but attention has to be paid on the a-priori information added to the ill-posed problem, implicitly by the algorithm or explicitly by the selected parameters. This requires extensive studies on the reliability of the reconstructed concentration fields (e.g., Laepple et al., 2004, Hartl et al., paper in preparation).

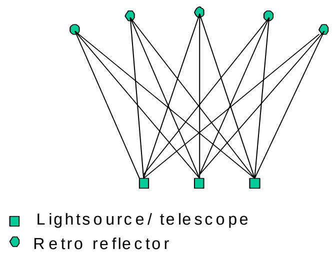

Figure 1. Measurement principle of a groundbased DOAS tomographic setup. Here, three telescopes emit five light beams each, which are reflected back by the retro reflectors.

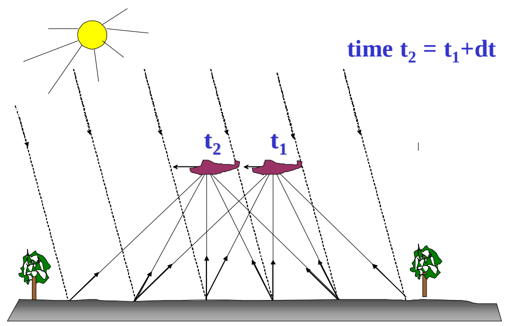

Figure 2. Measurement principle of an airborne tomographic setup. Here the light beams from five different directions below the aircraft are shown.

2. GROUNDBASED MEASUREMENTS

2.1. Measurements

The first tomographic arrangement (Fig. 3) was set up in the framework of a motorway emission campaign in April/May 2001 (Corsmeier et al., 2004). It consisted of two Longpath telescope sites and eight retro-reflector sites. Each telescope emitted one light beam, which was successively directed to the eight retro-reflectors (Pundt et al., 2005). The two light beams were reflected back to the telescopes and coupled into spectrometers. Thus 16 light paths were achieved in total.

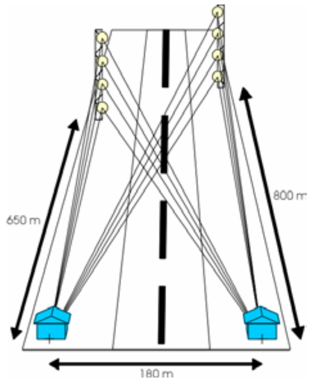

Figure 3. Tomographic setup during the motorway emission campaign. From each telescope one light beam is directed successively towards the eight retro reflectors (Pundt et al., 2005)

2.2 Tomographic reconstruction

Since the reflectors were pointed at successively, a tomographic reconstruction is possible only for stable meteorological conditions and a constant vehicle flux during one measurement cycle, which varied between 15 and 45 minutes. In Fig. 4, the two dimensional NO_2 concentration distribution perpendicular to the motorway is shown for 10 May 2001 (average from 0900 to 1300). For the inversion, the Simultaneous Algebraic Reconstruction Technique (SART) was used. The reconstruction is performed with bilinear basis functions on a 3 x 4 rectangular grid.

An emission plume from the location of the motorway towards the southern (right-hand) tower can be clearly seen in consistency with the wind direction measured during the presented period. The northern side represents the NO_2 background, and the southern side the emission plus the background.

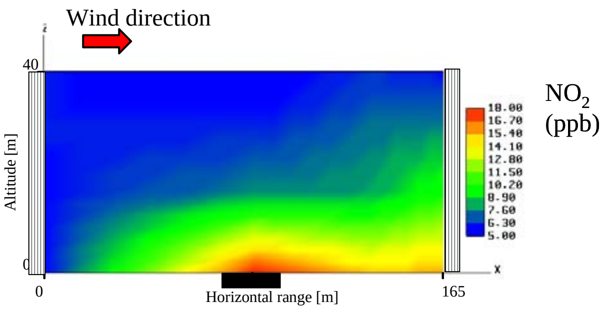

Figure 4. 2D reconstruction of the NO_2 emission plume (in ppbv) at right angles to the motorway, between the two towers (Pundt et al., 2005). The reconstruction is carried out over a four hour average in the morning on May 10th 2001 (0900-1300 CET). Black bar: the motorway, shaded areas: the two towers.

3. INSTRUMENTAL DEVELOPMENTS

Because of the limited time resolution given by the instruments employed above, a new Longpath DOAS telescope type, the “Multibeam telescope”, was developed for the simultaneous measurement along multiple paths (Pundt and Mettendorf, 2005). This telescope emits up to six light beams simultaneously, using only one single lamp as light source. Fig. 5 shows the instrumental setup. Here, two light beams are shown. They result from two ‘virtual lamps’ created by additional mirrors inside the lamp housing.

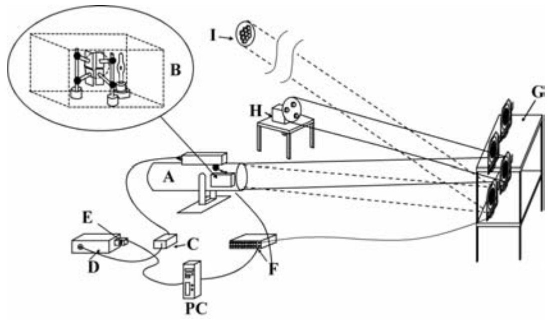

Figure 5. Instrumental setup of the novel Multibeam system including telescope A, lamp housing B, mode mixer C, spectrograph D, CCD detector E, stepper motor controller F, mirror tower with four mirror units G, the Shortpath reflectors H and the Longpath reflectors I (Pundt and Mettendorf, 2005).

Fig. 6 shows a picture of the Telescope taken from the front. Two virtual lamps can be seen on the telescope´s main mirror.

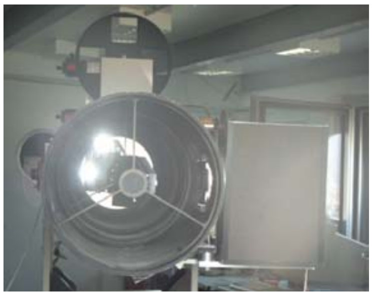

Figure 6. Picture of the Mulitbeam telescope from the front. Two virtual lamps appear on the telescope main mirror.

4. TOM-DOAS VALIDATION CAMPAIGN

An indoor validation experiment with specified concentration distributions was conducted in August 2003 in order to validate the measurement method (Fig. 7).

Figure 7. Picture of the Tom-DOAS validation experiment. One Multibeam-system (left), one polycarbonate cell (middle), and one mirror tower (right side) can be seen. Four light beams from another telescope are directed onto the experimentalists.

Above an area of 15m x 10m the atmospheric boundary layer was simulated in small scale. The setup corresponds to a small town with a ground surface of 1,5 km x 1 km containing two emission plumes with a diameter of 200 m. The emission plumes were simulated by one or two NO_2–filled cells, diameter of 2 meters each. The measurement geometry and one of the cells are displayed in Fig. 8.

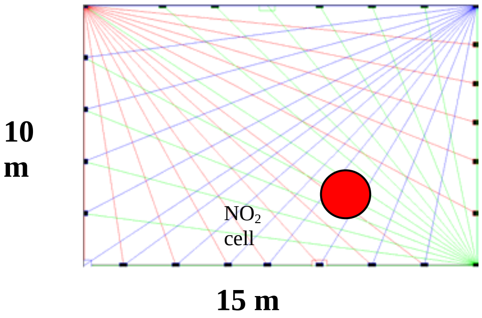

Figure 8. Measurement geometry of the Tom-DOAS validation experiment.

Then the NO_2 2D concentration distribution was measured using 3 telescopes, 12 simultaneous light beams, and 39 light paths in total. An example of a reconstructed concentration distribution is shown in Fig. 9. The comparisons between the given and the reconstructed distributions show very good agreement, proving the method valid for specific atmospheric conditions (Mettendorf et al., paper in preparation).

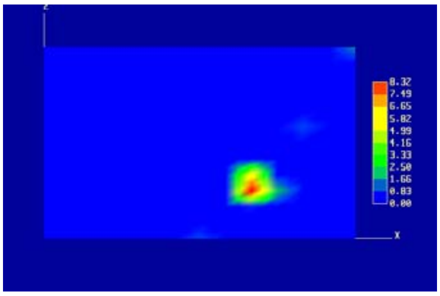

Figure 9. Tom-DOAS validation experiment: Reconstructed result from one data set.

5. AIRCRAFT MEASUREMENTS

5.1. Measurements

Multi-Axis DOAS instruments were installed in a small aircraft (Partenavia 68). The instruments consist of two spectrometers (covering the UV and visible spectral ranges, respectively) that are connected to ten telescopes (e.g. Pundt et al., paper in preparation). Three of the telescopes are pointing upwards under different angles, the remaining seven are pointing downwards (Fig. 10).

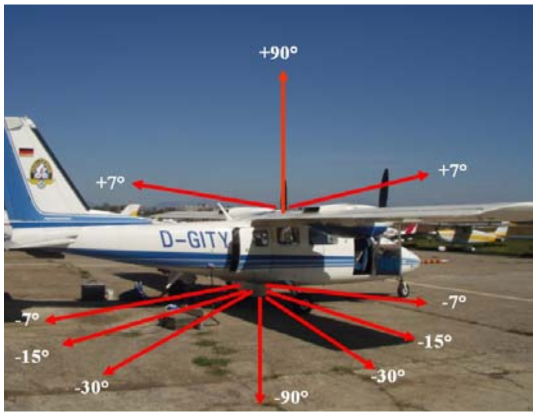

Figure 10. Partenavia Aircraft and the viewing directions of the telescopes during the second Format campaign in September /October 2003.

20 flights of about two hours each were performed in the Milan area (Italy) in the framework of the EU "FORMAT" project in summer 2002 and autumn 2003. Atmospheric spectra were recorded every 30 seconds corresponding to a horizontal resolution of about 2 km. Fig.11 shows the flight track on the 26 September 2003 together with the NO_2 slant columns derived with the spectral analysis.

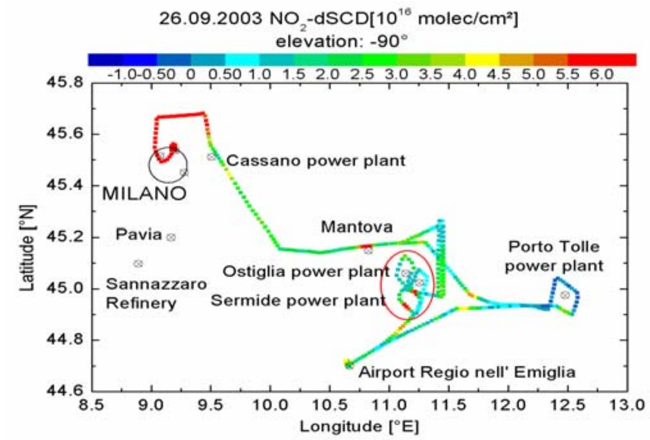

Figure 11. NO_2 slant column density as a function of the geographic position for the 26 September 2003. The power plant of Sermide, which is marked by the red circle, was surrounded and its plume was over flown three times

In Fig. 12 an optical sketch of the overlapping solar light paths is shown for downward looking telescopes for a plume measurement near the Sermide power plant. For clearness, only the light paths between the ground and the telescopes – assuming single reflection on the ground - are shown.

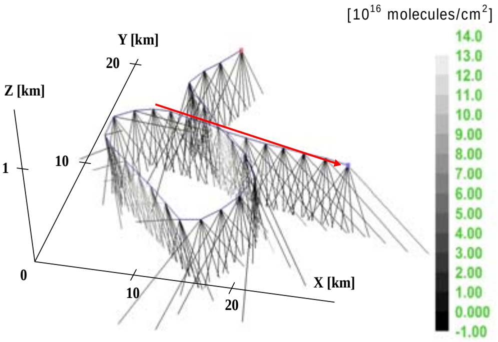

Figure 12. Aircraft measurement geometry near the Sermide power plant on 26 September 2003. Only the solar light paths from the ground towards the telescopes are shown for clearness. The intensity of the beams presents the measured NO_2 column density. The red flash marks the location and direction of the third overflow which is presented here.

5.2. Tomographic reconstruction

For the reconstruction, only the reflection on the ground was assumed. Fig. 13 displays the NO_2 map derived from one pass over the Sermide emission plume.

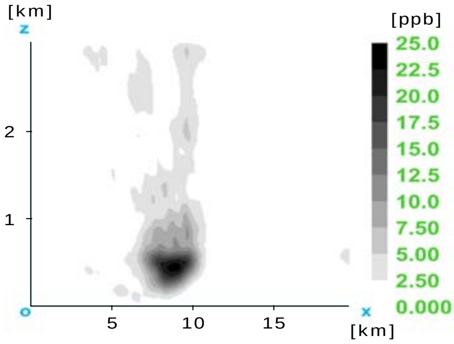

Figure 13. NO_2 2D mixing ratio distribution after tomographic inversion for the third overpass, performed at 5 km distance from the chimney.

6. SOFTWARE DEVELOPMENT

The experimental software TOMOLAB has been developed and used for the optimisation and inversion of tomographic measurements. It comprises different inversion techniques, and allows the choice of a large variety of parameters (e.g. basis functions, grid, and constraints). For theoretical studies, the software is able to calculate slant column data from CTModel concentration fields and assumed errors. The reconstruction results of these theoretical data are then evaluated by comparison with the original data (Laepple et al., 2004).

7. FUTURE CAMPAIGNS

Two-dimensional measurements of the horizontal distribution of SO_2, NO_2, HCHO and HONO are scheduled for spring-autumn 2005 above the city of Heidelberg (Germany). For details see Pöhler et al. (Instrumental setup and measurement configuration for 2D tomographic DOAS measurements of trace gas distributions over an area of few square km, this issue).

8. ACKNOWLEDGEMENTS

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