Instrumental setup and measurement configuration for 2D tomographic DOAS measurements of trace gas distributions over an area of a few square km

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LP-DOAS (Long path Differential Optical Absorption Spectroscopy) tomography allows determining 2 and 3 dimensional trace gas distributions by measuring the average concentration of different trace gases along 10 to 20 light paths and applying tomographic inversion techniques.
A configuration of three Multibeam Long path DOAS Instruments and almost 20 different light paths to retro reflector arrays will be used in spring 2005 for 2D measurements of NO_2, SO_2, O_3, HCHO, and HONO over an area of a few square km above the city center of Heidelberg. With each of the telescopes it is possible to measure along up to six light paths at once.
Later measurements at shorter wavelength will be performed to investigate the concentrations of the aromatic compounds Benzene, Toluene and X-Phenol.
The 2D trace gas distributions will be obtained using reconstruction techniques.

Keywords: DOAS, Tomography, trace gases, Multibeam Long path telescope

1. INTRODUCTION

LP-DOAS is a well known technique for measuring tropospheric trace gases (e.g. Platt, 1978). First LP-DOAS instruments consisted of an emitting and a receiving Newton telescope (Platt et al., 1979; Platt and Perner, 1984). They measure the average concentrations of the trace gases along the light path between the two telescopes. But single path measurements are often not sufficient (as well as single point measurements), if small-scale variations exists or transport is important. For this it is necessary to measure along a couple of light paths.
The application of tomographic techniques on outdoor measurements was recently investigated (Pundt et al., 2005; Laepple et al., 2004; Knab, 2003). It shows that the use of the necessary telescopes is too expensive and not practical because of low time resolution, so that a new type of telescope, the “Multibeam LP-DOAS telescope”, was developed to allow efficient measurements along up to six light beams simultaneously (Pundt and Mettendorf, 2005). With the suitable combination of two or more Multibeam LP-DOAS telescopes and the use of tomographic reconstruction techniques good quality 2D reconstructions of the measured trace gases can be achieved.
The obtained information’s from the experiment are not only helpful for pollution monitoring, but also to validate chemical transport models.

In this work we present the setup for 2D outdoor measurements over an area of 3 x 4 km^2 above the city centre of Heidelberg using tomographic DOAS technique.

1.1 Basic LP-DOAS principle

The earliest LP-DOAS setup uses a telescope to emit a light beam originating from an artificial light source (typically Xe-Arc lamp), which travels several 100 meters through the atmosphere, and which is then collected by a second telescope (Platt, 1978). The received light contains information’s of the trace gas absorptions, typically in the UV and visible wavelength range. Quantitatively the absorption of radiation is expressed by Lambert-Beers Law:

$$I(\lambda) = I_0(\lambda)\exp(-\int_0^L \sum_i \sigma_i(\lambda)\cdot c_i \cdot dl) \qquad (1)$$

Where $\sigma_i(\lambda)$ denotes the absorption cross section at the wavelength λ, while $I_0(\lambda)$ is the initial intensity emitted from the source and $I(\lambda)$ is the radiation intensity after passing through an air volume with a length L. Since atmospheric scattering and broad-band absorption can hardly be described, the absorption cross section σ is divided in two parts:

$$\sigma(\lambda) = \sigma_{broad}(\lambda) + \sigma'(\lambda) \qquad (2)$$

$\sigma'(\lambda)$describes only the characteristic narrow band absorption structures of different trace gases. For a DOAS setup Lambert-Beers Law can be expressed as:

$$I(\lambda) = I_0(\lambda)\exp(-\int_0^L \sum_i \sigma_{i,broad}(\lambda)\cdot c_i \cdot dl \times \exp(-\int_0^L \sum_i \sigma_i'(\lambda)\cdot c_i \cdot dl \qquad (3)$$

$$I(\lambda) = I_0'(\lambda)\exp(-\int_0^L \sum_i \sigma_i'(\lambda)\cdot c_i \cdot dl \qquad (4)$$

$I_0'(\lambda)$ contains the initial intensity and the broad band absorption structures and is computed from the measured spectrum $I(\lambda)$ by applying a low pass filter. The differential absorption cross sections $\sigma_i'(\lambda)$ have to be treated with the

same high pass filter algorithm from in laboratory measured absorption cross sections $\sigma_i(\lambda)$.
With this technique it is possible to determine many different trace gases simultaneously like O_3, SO_2, NO_2, HONO, HCHO, BrO, NO_3, ClO, IO (Platt, 1978, 1994) and the BTX aromatic compounds Benzene, Toluene and X-Phenol (Volkamer et al. 1998; Ackermann 2000).
With the use of the coaxial telescope (Figure 1) it was possible to transmit and receive the light beam with one telescope (Axelsson et al., 1990). With this telescope the emitted parallel light beam is reflected after some 100 meters back into the telescope.

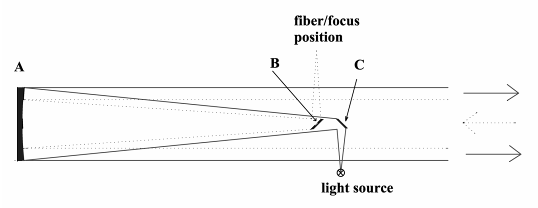

Figure 1. The coaxial Newton telescope design of Axelsson et al. 1990

Cube retro reflectors are used, because of their property to reflect the light exactly back into the direction of incidence, independent of their exact orientation. Therefore the light from the telescope will be exactly reflected back into the telescope, and the adjustment is much easier.

1.2 Long path DOAS Tomography

Long path DOAS Tomography is a combination of multiple Long path DOAS measurements with tomographic inversion techniques. 10 to 30 or even more overlapping or non overlapping light beams are used to probe the concentration field from different directions (Pundt, this issue; Laepple, 2004; Hashmonay et al., 1999; Price, 1999; Hartl, this issue).
The successive scanning of all light paths with normal Long path DOAS instruments is not suitable because of the low time resolution. With the use of Multibeam Long path DOAS instruments and measuring up to six light beams with each telescope simultaneously, the time resolution can be enhanced by a factor of 10 to 100.

1.3 Multibeam LP-DOAS

For tomography the system described in section 1.1 has been modified to a Multibeam LP-DOAS telescope which emits several light beams with small angles to each other (Pundt and Mettendorf, 2005). Figure 2 shows a sketch of a complete system with four emitting beams.
The telescope **A** has a main mirror of f=1500mm and 300mm diameter and is similar to a coaxial Newton telescope. The crucial change was realized inside the lamp housing **B**. Up to six small mirrors (tree in vertical and two in horizontal direction) inside the lamp house produce each one virtual light source in the focal plane of the main mirror. Each virtual light source produces an emitting telescope light beam in a specific direction, depending on the position and orientation of the lamp housing mirror. For 6 beams the maximum diverging angle to each other is 1.2° vertical and 1.5° horizontal. An existing telescope with such lamp housing is shown in figure 3.

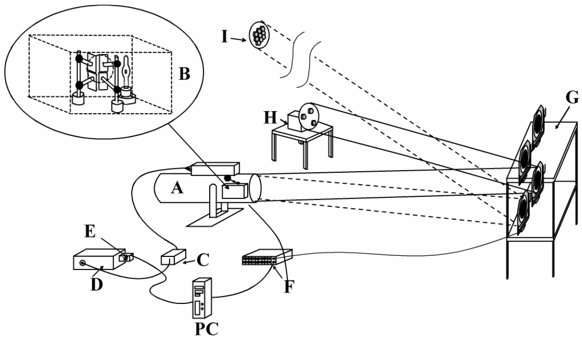

Figure 2. Experimental setup of an entire Multibeam system (Pundt and Mettendorf, 2005)

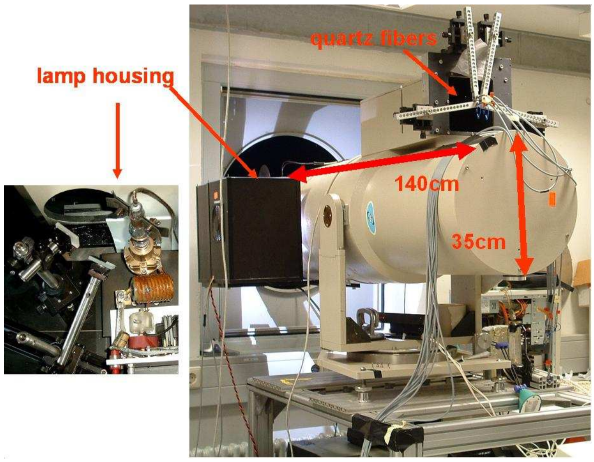

Figure 3. Multibeam Long path DOAS instrument (Hydra). Extended view: inside of the lamp housing from above

In front of the telescope plane mirrors (360 x 320 mm^2) are positioned on the tower **G**. A distance of 24 meters is necessary to separate six beams enough for the plane mirrors incl. frame.
For shorter distances only fewer beams can be used. The plane mirrors redirect each beam into the desired direction, for example to a retro reflector array **I** far away or to a rotating retro reflector array **H** close by. The mirrors can be turned in horizontal and vertical direction with stepper motors by the controller **F**. After reflection at the retro reflectors the light is sent back to the tower mirrors and from there to the telescope. For each beam the received light is focused on a quartz fiber and than sent over a mode mixer **C** (Stutz, 1996) to an imaging Czerny-turner spectrograph where a 2D CCD chip **E** measures the spectrally analyzed signal. (Pundt and Mettendorf, 2005)
For DOAS measurements usually a lamp reference or correction spectrum is required which accounts for the shape of the lamp spectrum and absorption structures given by the instrument. For a shortcut spectrum the light beam is directed to a close short cut retro reflector array **H.** The array is rotating to laterally distribute the reflecting surface as homogeneously as possible.

2. THE MEASUREMENT SETUP OF HEIDELBERG

An experiment will start in spring 2005 to measure the concentration distribution above the city centre of Heidelberg, Germany. The setup consists of 3 Multibeam telescopes located on top of high buildings in the city and almost 20 retro reflector arrays, which are mounted on steeples, typically.

2.1 The instruments

Three Multibeam Long path DOAS instruments as described in 1.2 are used for the experiment. One mirror tower by night is shown in figure 4.

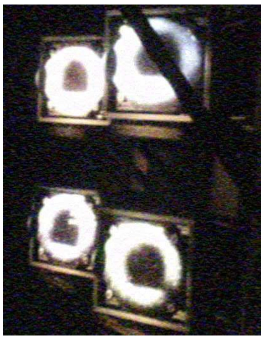

Figure 4. Tower mirrors by night which redirect the light beams towards the retro reflector arrays

The beams are redirected to different retro reflector arrays at different distances. The number of used cube retro reflectors for one array varies strongly with the measured distance from 7 up to 20 reflectors.

2.2 The measurement geometry

The measurement is performed over an area of 3 x 4 km². The geometry is displayed in figure 5.

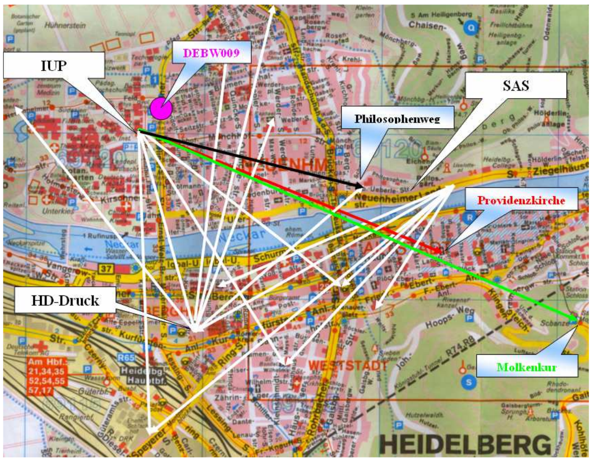

Figure 5. Measurement geometry in Heidelberg with three Multibeam telescopes and highlighted paths of the first measurements

Three telescopes are located on top of the buildings “IUP” (Institute of Environmental Physics), “SAS” (SAS Institute GmbH, Germany) and “HD-Druck” (Heidelberger Druckmaschinen AG), from where the beams are send to different retro reflector arrays. The number of light paths is limited by the mountains north and south of the Neckar River. In the realized geometry all possibilities were utilized to achieve a regular covering with 19 partly overlapping beams in one height.

Theoretical studies use other geometries with more light beams in regular arrangements to reconstruct concentration distribution (Hartl et al., this issue). But these geometries are mostly just realizable in indoor experiments like made by (Mettendorf et al., paper in preparation). Due to differences to this geometry the reconstructed 2 dimensional trace gas distribution will have a lower resolution.

2.3 Measurable trace gases

Typical measurements are performed over a wavelength range from 295 to 375 nm and allow the reconstruction of O_3, SO_2, NO_2, HONO and HCHO in Heidelberg from one measurement simultaneously. The spectrograph gives also the possibility to measure at shorter wavelength range and allows measurements of the aromatic compounds Benzene, Toluene and X-Phenol in additional measurements.

2.4 Automatic measurement

All measurements which are necessary for a LP-DOAS analysis are taken automatically. In regular intervals the aperture records different measurements of: background spectra from light which is scattered into the telescope from other light sources; lamp spectrum over the shortcut retro reflector array; mercury lamp spectra to correlate the channel on the CCD chip to a wavelength which can change with the time and additional measurements to correct measurement errors of the CCD chip.

3. FIRST MEASUREMENTS

First measurements of NO_2, SO_2, O_3, HONO and HCHO have been performed in December 2004 with the instrument on top of the “IUP” building to three different reflector positions: “Philosophenweg”, “Providenzkirche” and “Molkenkur” (see figure 5). The telescope and the reflectors “Philosophenweg” and “Providenzkirche” are in one layer and the reflector “Molkenkur” is located on a mountain. Derived NO_2 average mixing ratios from these light paths are shown in figure 6a) and additionally the measured concentrations from a point measurement “DEBW009” are plotted in the same diagram (B. Rippel, Diploma thesis in preparation). Apparently the beam to the “Providenzkirche” measures higher concentrations than the beam to the “Philosophenweg”, the first crosses more high traffic. The higher altitude beam to the “Molkenkur” reflector measures lower concentrations due to the vertical dispersion of NO_2. After tomographic inversions these differences in mixing ratios would yield in non homogeneous concentration distribution. The high differences show that tomography on this scale is possible. A general correlation to the in situ measurement “DEBW009” can be seen. Differences occur due to the measurement near a high traffic road. In Figure 6b) with a seven hour zoom additionally temporal differences are visible which can be described by transport models.

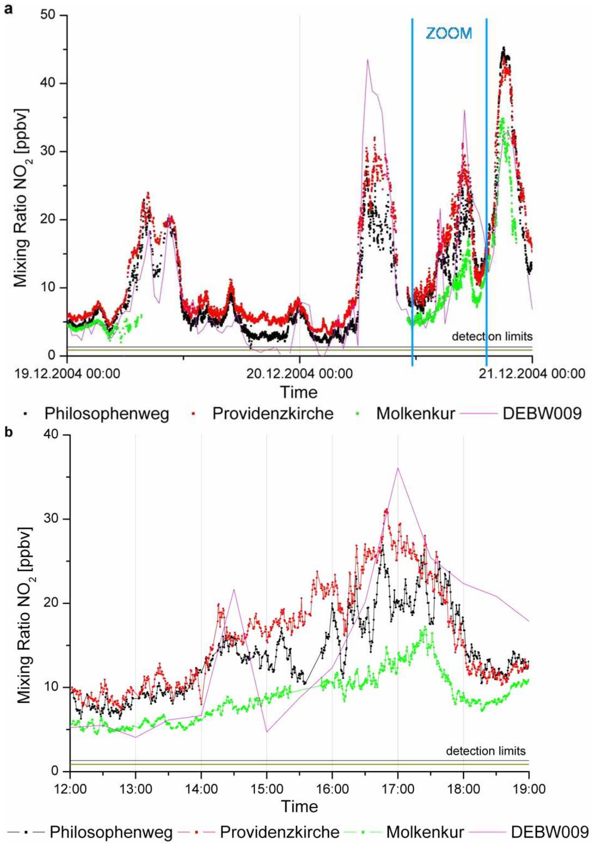

Figure 6a) NO_2 average mixing ratio measured during two days along three different light paths together with results from in situ measurement "DEBW009"; b) 7 hours zoom from plot a)

4. Outlook

After the installation of all telescopes and retro reflectors it will be possible to run the system continuously over a time scale of several months. Finally the tomographic inversion techniques will be applied to obtain the trace gas concentration distributions as function of time.

With the installation of retro reflector arrays on the mountains north- and southeast from the city centre additional measurements into the upper layer will be possible and will allow tomographic reconstruction in 3 dimensions.

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6. REFERENCES

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