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A multi-path differential optical absorption spectroscopy system for urban pollution monitoring

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ABSTRACT

A fully computer-controlled differential optical absorption spectroscopy (DOAS) system for atmospheric air pollution monitoring is described. A receiving optical telescope can via a large planar mirror controlled by stepping motors sequentially tune in to light beams from a number of distant lamp light sources to cover the area. The light is directly coupled into a rapid scanning spectrometer from the computer-controlled secondary mirror in the Newtonian telescope. The beam-finding servo system and automatic gain control allows long unattended measurements. Using an astronomical code, celestial sources can also be searched and tracked. By computer fitting to stored laboratory spectra the path-averaged concentration of a number of important pollutants such as NO_2 , SO_2 and O_3 can be evaluated. A measurement of NH_3 and NO close to the UV-limit is also demonstrated. Evaluated data are stored together with meteorological data and other system parameters.

1. INTRODUCTION

With the growing awareness of the serious environmental impact of industrial and automotive operations the need for the development of powerful measurement techniques for atmospheric air pollutants is increasing. Optical remote sensing techniques using lidar (light detection and ranging) and long-path absorption methods are particularly advantageous allowing large-area monitoring and avoiding sample preparation difficulties characteristic of many point monitoring techniques.¹⁻³ The differential absorption lidar (DIAL) technique, using powerful pulsed laser sources, allows a three-dimensional mapping of the atmospheric pollutants, while cw laser sources provide a very high spectral resolution and sensitivity in long-path absorption applications. Systems using classical light sources combined with optical spectroscopy in differential optical absorption spectroscopy (DOAS) or IR-light sources combined with Fourier transform spectroscopy (FTIR), do not have the very high performance of laser-based systems but, on the other hand, they can provide realistic, automated and enduring measurement capabilities for multi species monitoring as the DOAS system demonstrated in this paper.

The DOAS technique was pioneered by Platt, Perner and co-workers.⁴⁻⁶ A number of species have been measured, see e.g. Ref. 6. In the present paper we describe a DOAS monitoring system allowing automated monitoring of the air mass above a medium-sized city and with some specialised research capabilities.⁷ The hardware description of the multi-path atmospheric spectroscopy system is given in Sect. 2.1, while measurement routines are described in Sects. 2.2. Atmospheric air pollution spectroscopy performed with the system and practical concentration evaluation are illustrated in Sect. 3 for SO_2 , NH_3 and NO . Examples of data collected with the system over a period of time are given in Sect. 4.

2. SYSTEM DESCRIPTION

2.1 Hardware

A schematic diagram describing the DOAS system is shown in Fig. 1. The light source normally used for DOAS is a high-pressure xenon short-arc lamp. The lamp is placed at the focus of a spherical mirror to obtain a well collimated light beam. The xenon lamp has a broad, smooth emission spectrum except for some regions with strong emission lines in the blue and near infrared regions.

The vertically positioned Newtonian receiving telescope has a primary mirror of 30 cm diameter. In front of this there is a large planar mirror, which can be rotated horizontally and vertically by stepping motors (0.47 mrad/step and 0.63 mrad/step, respectively), enabling the telescope to look in different directions. To improve the angular resolution of the telescope orientation the secondary mirror is controlled by stepping motors. This unit has a resolution of 0.025 mrad/step.

The dispersive instrument is a Spex 500 M spectrometer (f/4) with a 1200 groove/mm grating with blaze at 300 nm. A slotted disk scans the spectrum in 10 ms. The disk has twenty slits of 100 μm width and rotates at 5 rev/s. A mask in the focal plane limits the scan to cover approximately 40 nm. A trigger signal, generated when the slit passes an infrared light barrier close to the edge of the mask, is used to synchronise the wavelength scale of each individual scan. The entrance slit of the spectrometer is normally 100 μm wide which gives an optimum spectral resolution of 0.23 nm. The detector is an EMI 9558 QA photo multiplier tube (PMT) with a 5 cm diameter photo-cathode to capture the light passing the scanning exit slit. After amplification the signal is stored in a custom made multichannel analyser (MCA) plug-in card in an IBM-compatible AT computer. The MCA is equipped with a 12-bit analogue-to-digital converter (ADC) which divides each scan into 1024 channels and can add up to 60 000 scans before the result is transferred to the computer.

Presently, the system is arranged with three measurement paths overlooking the city of Lund, Sweden (pop. 50 000). Three individual xenon lamps are placed 2000 m, 1600 m and 400 m, from the receiver. The optimum path length varies for different species. A trade-off has to be made between higher sensitivity for longer paths and higher transmission for shorter paths which are less likely to be blocked by fog. The light paths are located at a height of 10 to 30 meters above the ground. The diameters of the lamp telescope mirrors are 30, 15 and 30 cm and the lamps have output powers of 500, 75 and 150 W. Normally, the lamp telescopes are sealed from the atmosphere by a quartz window for protection.

2.2 Measurement routines

In a measurement sequence the telescope is first turned to the first specified light source and the grating is set for the appropriate wavelength region for the first species to be measured. The voltage of the PMT tube is set to an empirical value depending on the distance to the light source as well as its spectral power. Due to thermal effects and backlash in the stepping motor machinery, the viewing direction of the telescope is optimised by adjusting the secondary mirror with stepping motors. The PMT voltage is then regulated so that the signal from the interesting part of the spectrum corresponds to about 75 percent of the maximum level for the AD converter. After each measurement meteorological data (wind speed and direction) from a small weather station are measured and stored. While the MCA card is accumulating scans, the computer is free to evaluate the previous measurement, print the results and store them in the computer and/or on floppy disks. The results and all relevant data are stored in files in a format suitable for use in graphic presentation programs. After the measurement of a particular wavelength region is completed, a new region can be chosen to study different pollutants. Other light sources can then be selected.

Several celestial objects can be searched and tracked using an astronomical code for steering of the telescope.^{8,9} The use of the sun, moon and scattered sky light together with fixed light sources allow measurements of both the tropospheric and stratospheric concentrations of many species,¹⁰ but this option will not be demonstrated in this paper.

3. ATMOSPHERIC AIR POLLUTION SPECTROSCOPY

The atmospheric spectra are evaluated according to the algorithms described by Platt and Perner.⁶ Fig. 2 shows an example of a multi-species measurement at short UV wavelengths. An additional measurement path of 350 m was used and the receiving telescope was optimised for the short UV region. In Fig. 2a the atmospheric spectrum (thick line) is shown together with a reference spectrum of sulphur dioxide (SO_2) scaled to represent 5.0 $\mu\text{g}/\text{m}^3$ over the 350 m path (thin line). The atmospheric spectrum after subtraction of the SO_2 reference spectrum is shown in Fig. 2b together with an ammonia (NH_3) reference spectrum of 0.7 $\mu\text{g}/\text{m}^3$. The residual after subtraction of the NH_3 reference is shown in Fig. 2c together with the reference spectrum of nitric oxide (NO) scaled to equal 2.8 $\mu\text{g}/\text{m}^3$. The final residual is shown in Fig. 2d. The larger structures might be related to other species in the atmosphere or due to artefacts from the normalisation to the polynomial fit. The noise in this measurement, $\sim 1.5 \cdot 10^{-3}$, is quite large due to the small light flux in this wavelength region. In measurements of NO_2 around 430 nm the noise is an order of magnitude smaller. The detection limits for this 350 m path length are estimated to 0.4 $\mu\text{g}/\text{m}^3$ for NH_3 , 1.1 $\mu\text{g}/\text{m}^3$ for SO_2 and 1.7 $\mu\text{g}/\text{m}^3$ for NO. Multi-species spectra are normally evaluated with a simultaneous correlation of the reference spectra. Fig. 2 shows the contribution from the separate species and a sequential evaluation can be used if the correlation between the different reference spectra is small.

4. EXAMPLES OF RESULTS

Fig. 3 shows the concentration of ozone (O_3) and nitrogen dioxide for one day. The anti correlation of the O_3 and NO_2 concentrations due to atmospheric chemistry is clearly seen. Data for nitrogen dioxide in three directions recorded during 24 hours are shown in Fig. 4. The average concentration of sulphur dioxide as a function of wind direction, measured between the 29th of May and the 14th of July 1990 is shown in Fig. 5. The impact of winds from the south and south-east is clearly indicated.

5. DISCUSSION

Detection techniques based on optical spectroscopy have many advantages compared with conventional wet chemistry instrumentation. Primarily, a mean value over a long distance is often more representative than a value from a point monitor. A large area can be monitored from one station as shown in this study. Optical instruments can often provide rapid measurements giving values with high temporal resolution. A further advantage is the low frequency of calibration necessary. Spectroscopic instruments are also easy to computerise and DOAS systems can run unattended for long periods, communicating with the operator via a modem on the telephone network for automated data reports or reprogramming. An alternative approach to the one described in this paper is to replace the lamp telescopes with retro-reflectors and to construct the transmitting lamp and receiving telescope as one unit.¹¹ The DOAS system in Lund will be rebuilt this way. The practical and economical benefits of replacing active lamp telescopes with passive retro-reflectors encourage the use of a larger number of retro-reflectors which would ensure optimum path lengths for all meteorological conditions. Rain does normally not effect the measurements if the windows are shielded, but fog can block the light if the path is long. Haze can occasionally scatter sun light into the receiving telescope but this effect can be compensated for in the retro-reflector concept; the lamp is blocked while the scattered sun light is measured and this signal can then be subtracted from the measurements.

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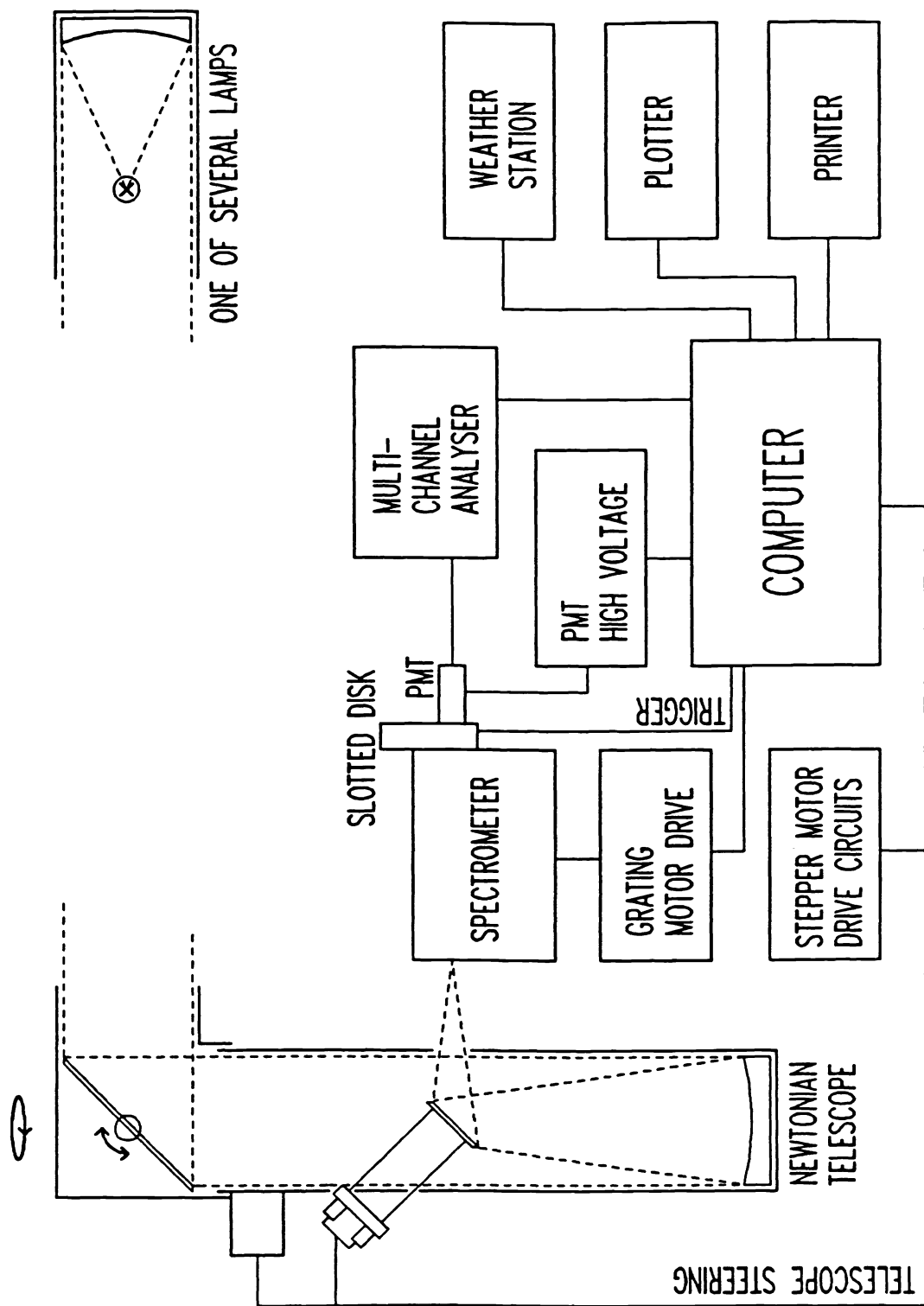


Fig. 1. Set-up of the DOAS station

(a) atmospheric spectrum (thick line)
and reference spectrum of $5.0 \mu\text{g}/\text{m}^3$
 SO_2 (thin line)

(b) atmospheric spectrum after SO_2
subtraction (thick line) and reference
spectrum of $0.7 \mu\text{g}/\text{m}^3$ NH_3 (thin line)

(c) atmospheric spectrum after SO_2 and
 NH_3 subtraction (thick line) and reference
spectrum of $2.8 \mu\text{g}/\text{m}^3$ NO (thin line)

(d) residual spectrum

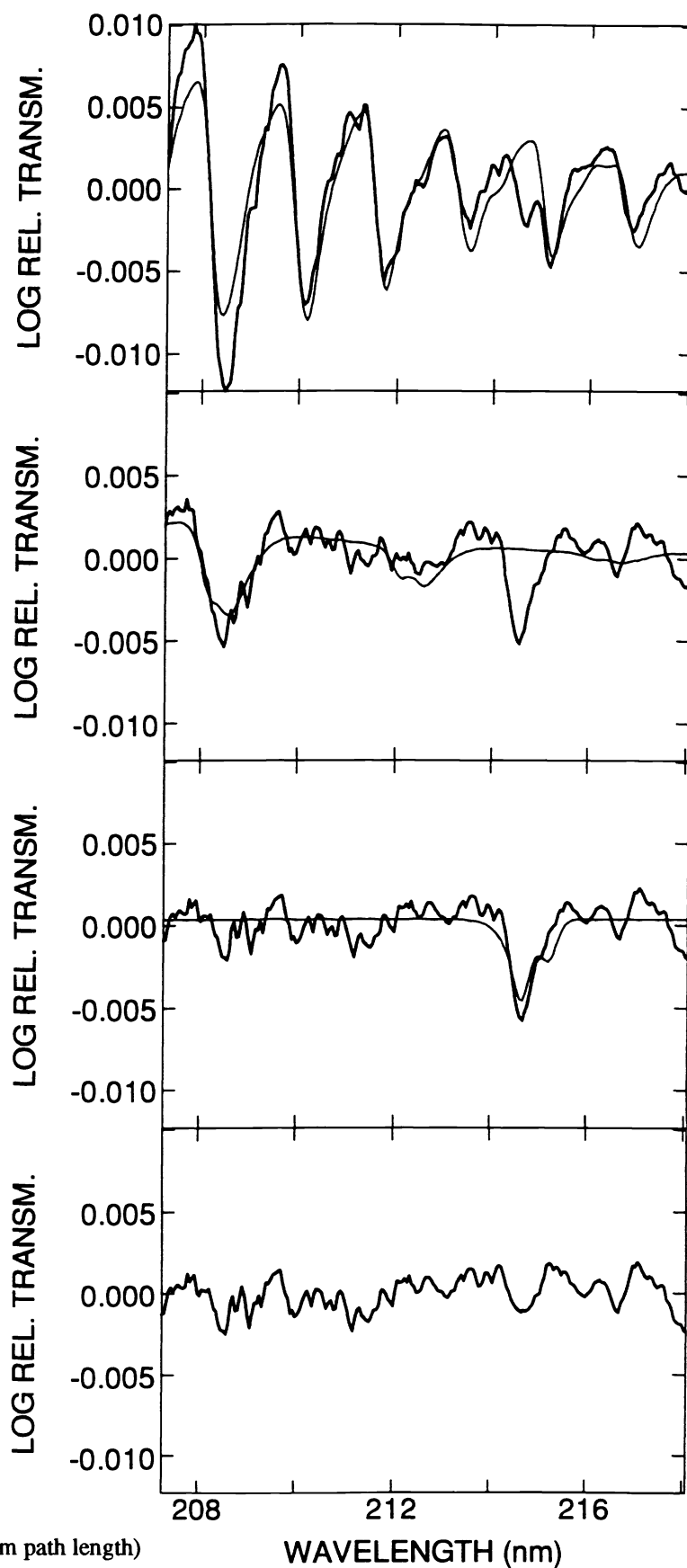


Fig 2. Measurement at short UV wavelengths (350 m path length)

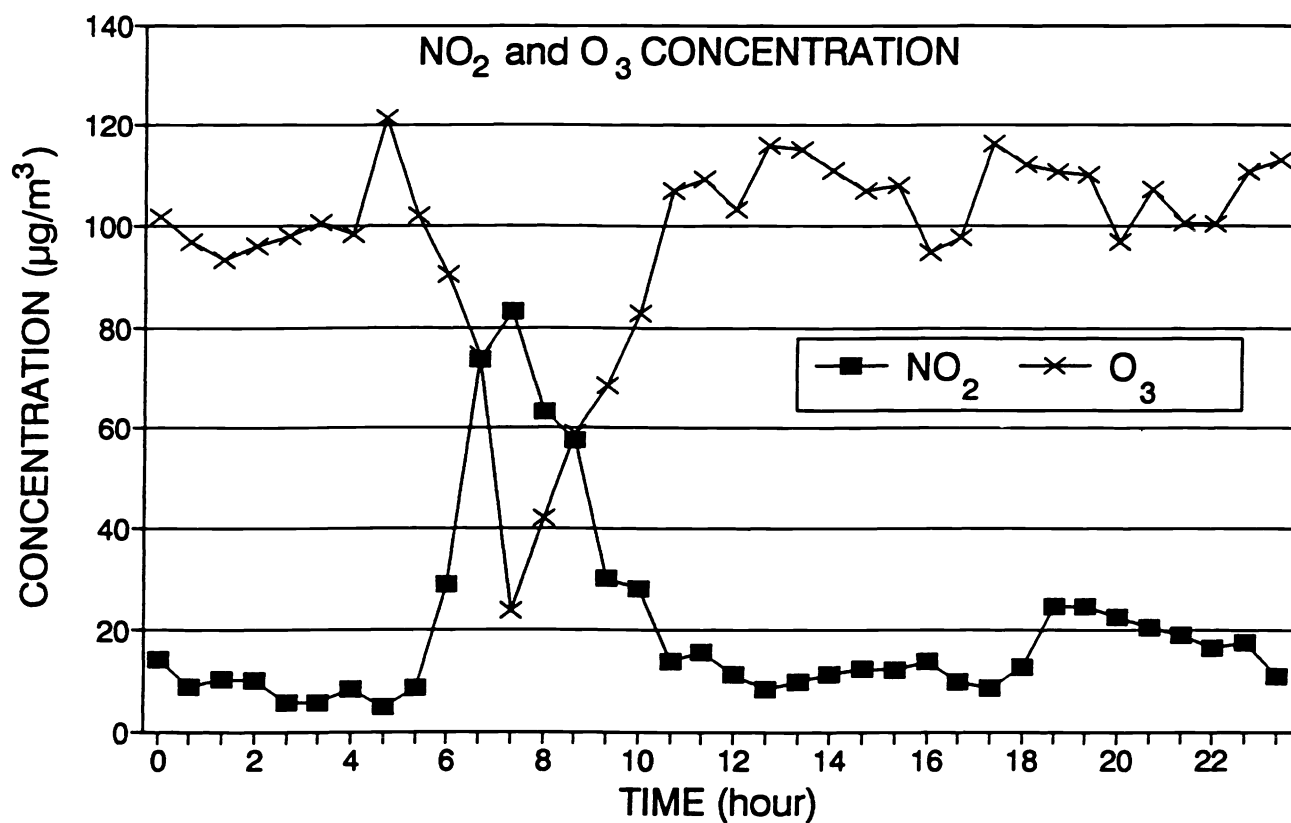


Fig. 3. NO₂ and O₃ correlation measured for 24 hours (400 m path)

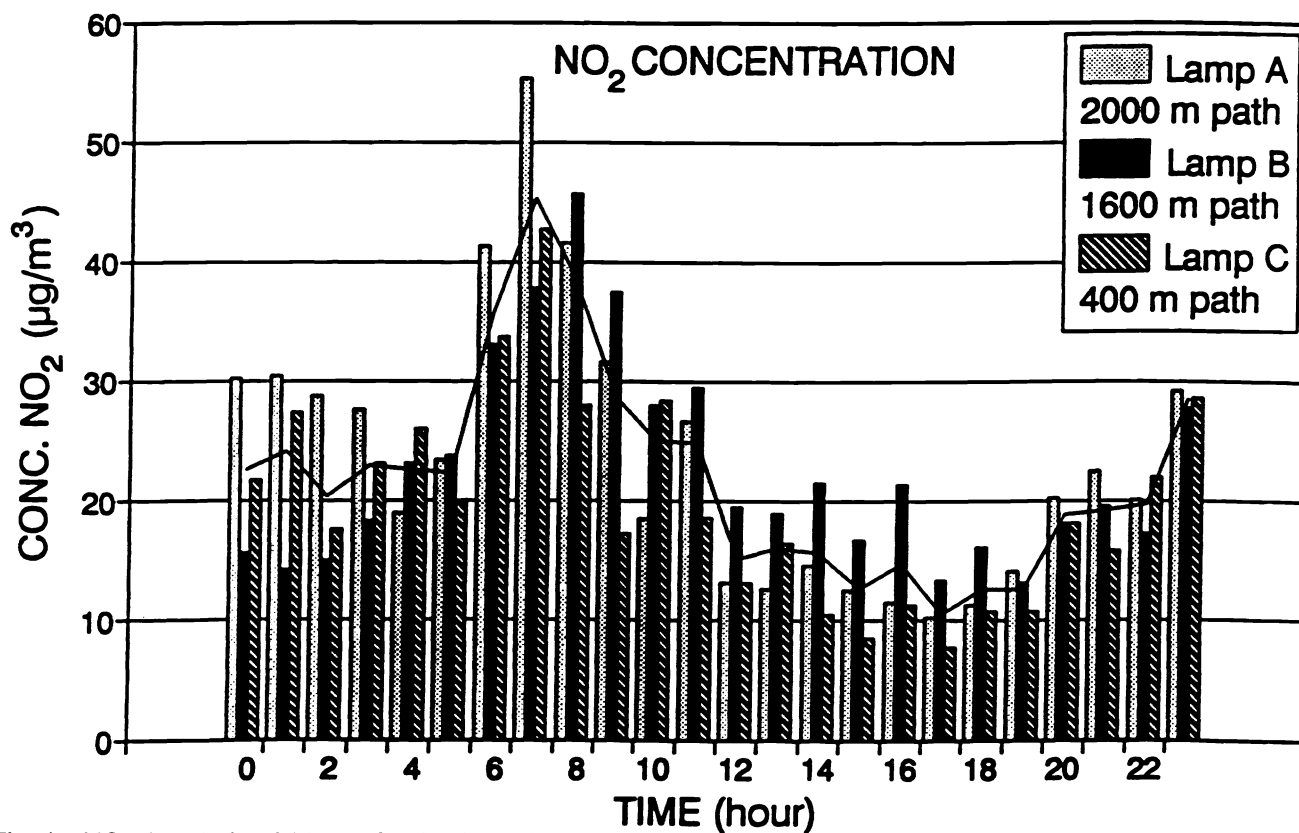


Fig. 4. NO₂ data during 24 hours for the three directions.

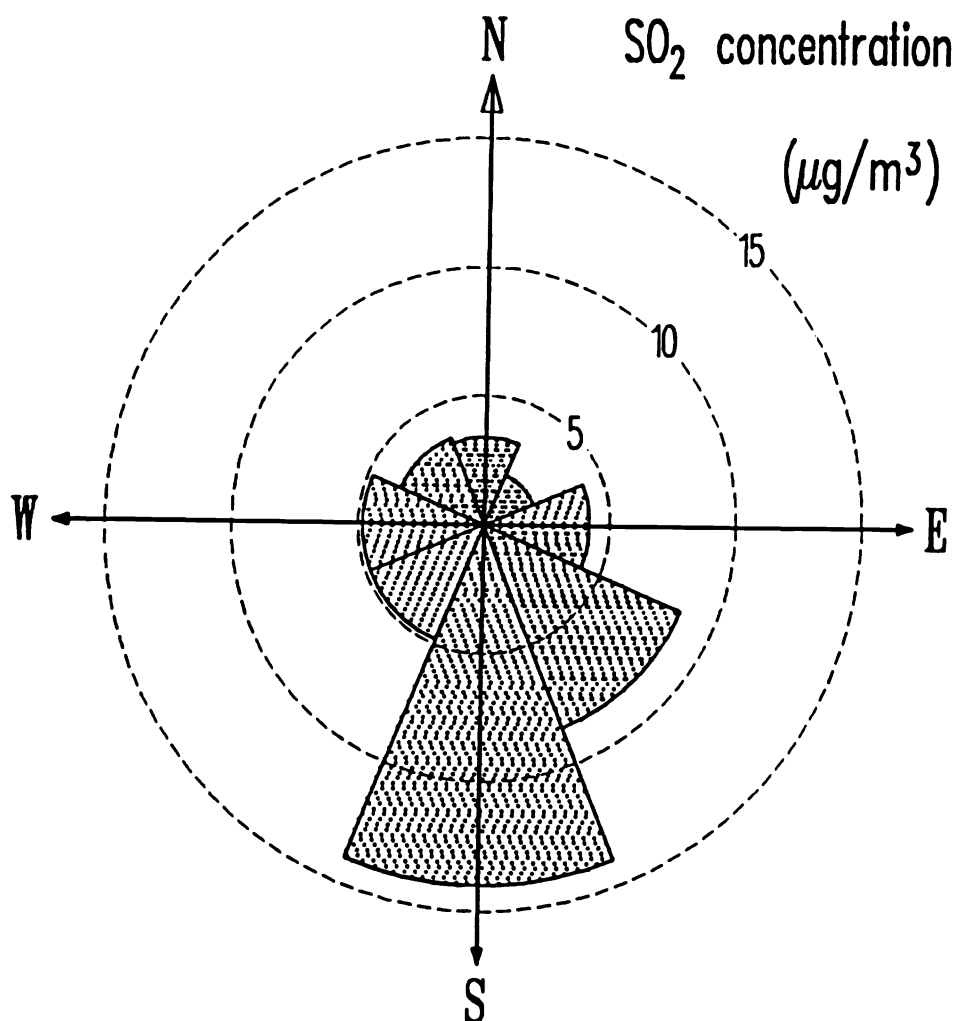


Fig. 5. Average SO_2 concentrations as a function of the wind direction, May 29 to July 14 1990 (2000 m path)