Measurements of NO_2 using MAX-DOAS observations of sun-illuminated targets

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Motivation & Physical background

Nitrogen oxide radicals (NO + NO2) are important trace gases in the atmosphere. They originate from combustion processes, lightning and soil emissions and largely control the tropospheric ozone production Since several decades it is possible to measure NO_2 with different techniques. However, for the spatial distribution in the troposphere considerable uncertainty exists. Here we present measurements of NO_2 on and close to the campus of the University of Bremen applying a novel technique: Topographic Target Light scattering Differential Optical Absorption Spectroscopy (ToTaL-DOAS, Frins et al., 2008). This experiment should provide a linkage between established methods like *in situ, MAX-DOAS* and satellite observations of NO_2.

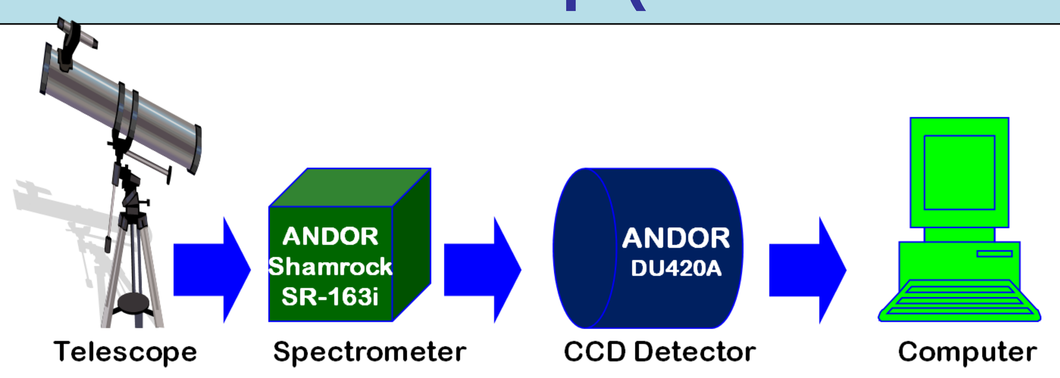
Idea:

•To collect sunlight reflected from natural and artificial targets (e.g. high buildings) at different distances from the measuring device
• Recorded spectra are analyzed for NO_2 applying the DOAS method (see below)
• Simple geometric treatments of the light-path reveal NO_2 concentrations in the boundary layer as a final data set.

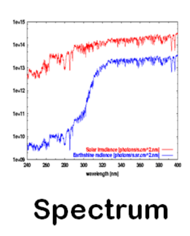
DOAS:

• Remote sensing measurement of atmospheric trace gases in the atmosphere
• Measurement is based on absorption spectroscopy in the UV and visible wavelength range following Lambert-Beer's law
• To avoid problems with extinction by scattering or changes in the instrument throughput, only signals that vary rapidly with wavelength are analyzed (thus the differential in DOAS)
• Measurements are taken at moderate spectral resolution to identify and separate different species

Instrumental Setup (Mobile MAX-DOAS) and Site



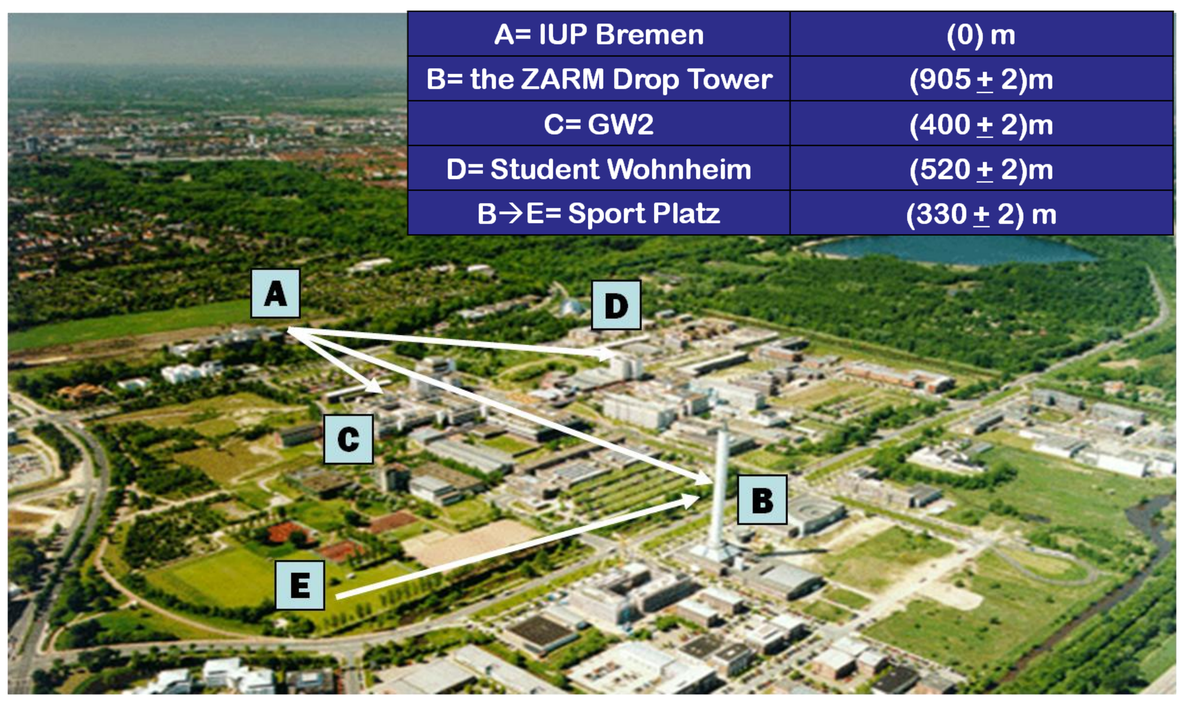
Detector Temperature	-34 ° C
Exposure time	0.400 sec
Integration time	60.0sec
Peak QE	95%
Wavelength window	422-- 510nm
Grating	1200 l/mm
Slit window	100µm
Focal Length	163mm
Wavelength Accuracy	± 0.25*(nm)



For the ToTaL-DOAS measurements a mobile DOAS setup (see scheme and technical parameters to the left) was modified using an astronomical mounting and a video camera. The latter is used to thoroughly pointing the telescope towards the different targets (see photo to the right). The correct alignment between video camera and the field of view of the telescope was checked by direct sun measurements.

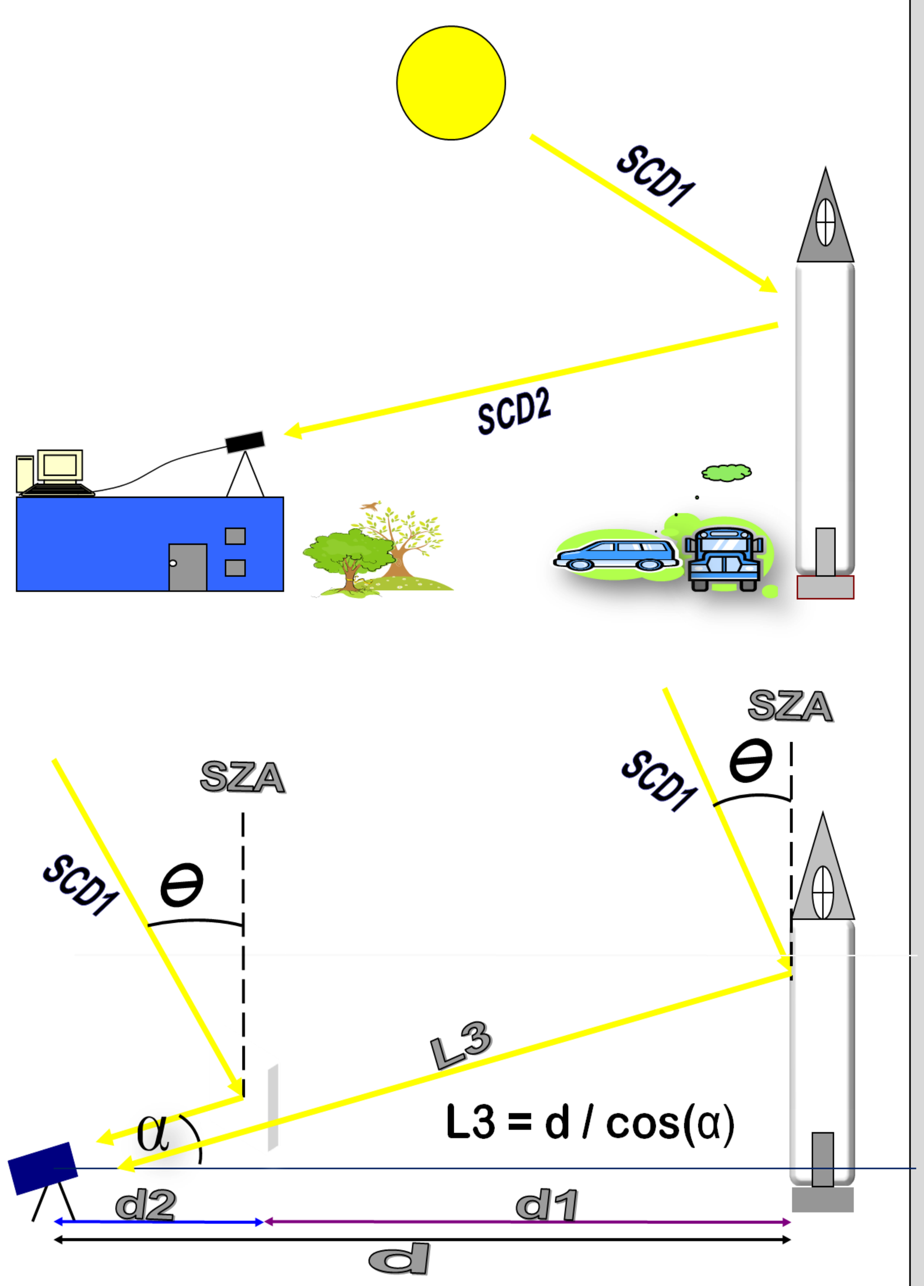
Most of the measurements were carried out from the roof of the iup building (A) . Between sites A and C, D there is a wide and busy road. Another highway is located in front of the drop tower (target B).

A= IUP Bremen	(0) m
B= the ZARM Drop Tower	(905 ± 2)m
C= GW2	(400 ± 2)m
D= Student Wohnheim	(520 ± 2)m
B→E= Sport Platz	(330 ± 2) m



Measurement Geometry for ToTaL-DOAS

ToTaL-DOAS means pointing a passive DOAS instrument at targets which are illuminated by direct and scattered sunlight. Applying the DOAS analysis to the recorded spectra one retrieves the slant column density (SCD, the integrated trace gas concentration along the light path, see upper sketch). In order to retrieve the NO_2 concentration between instrument and target one is interested into the partial column SCD2. The removal of the partial column between target and the top of atmosphere (SCD1) can be achieved recording a reference spectrum when the telescope is directed towards a target close to the instrument (lower figure). Ideally this target should have similar optical properties as the real target. Here a plate of styrofoam was used which was found to have the same characteristics as white painted concrete.



Vertical Scanning of a Drop Tower

The vertical scanning of a building or tower with the ToTaL-DOAS instrument has the potential to offer information about the vertical gradient of a trace gas. Here, for two days it was possible to scan the drop tower, a 146 m tall concrete shaft on the University of Bremen campus. Both data sets show decreasing NO_2 concentrations with increasing light paths (altitudes) which supports the hypothesis that the NO_2 is not well-mixed within in the boundary layer.

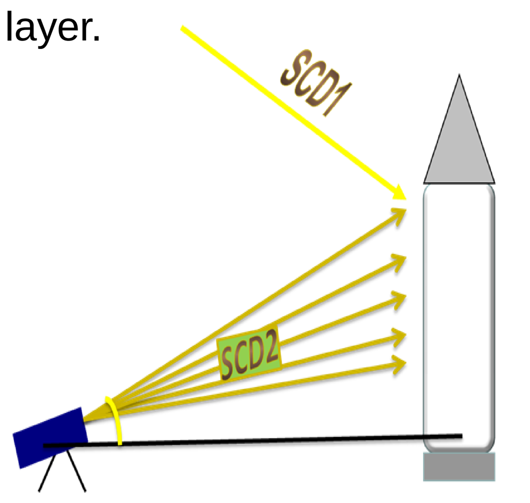
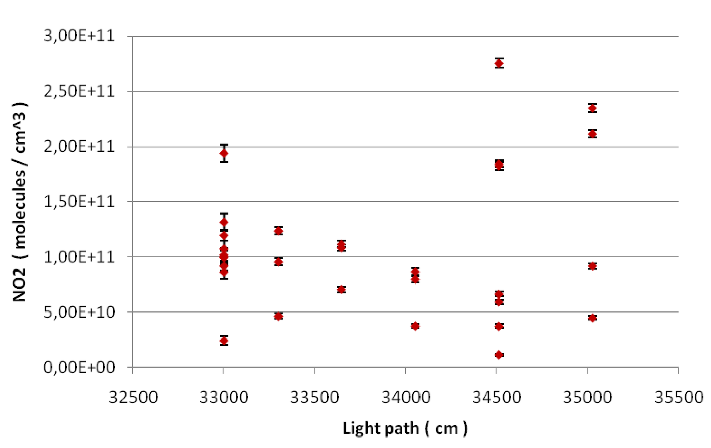
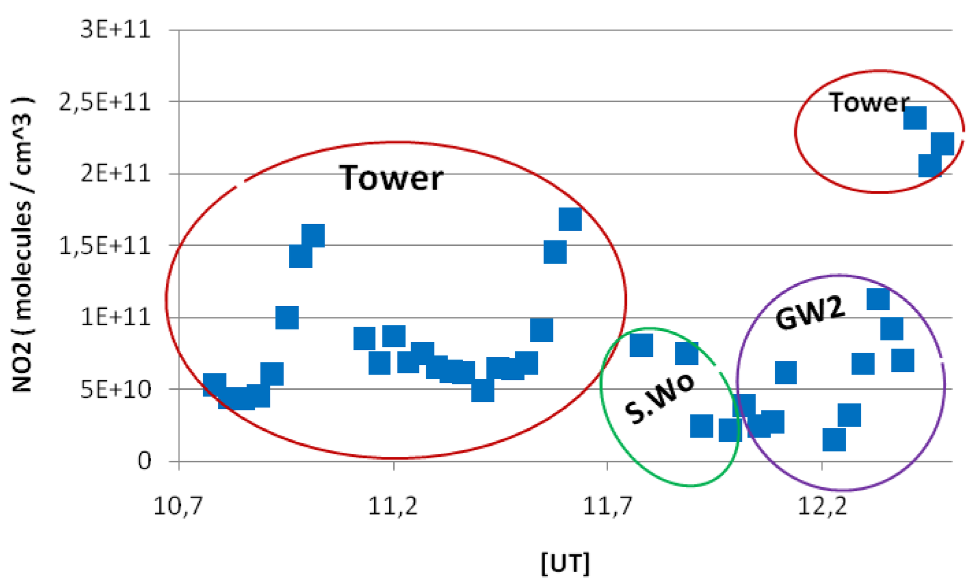


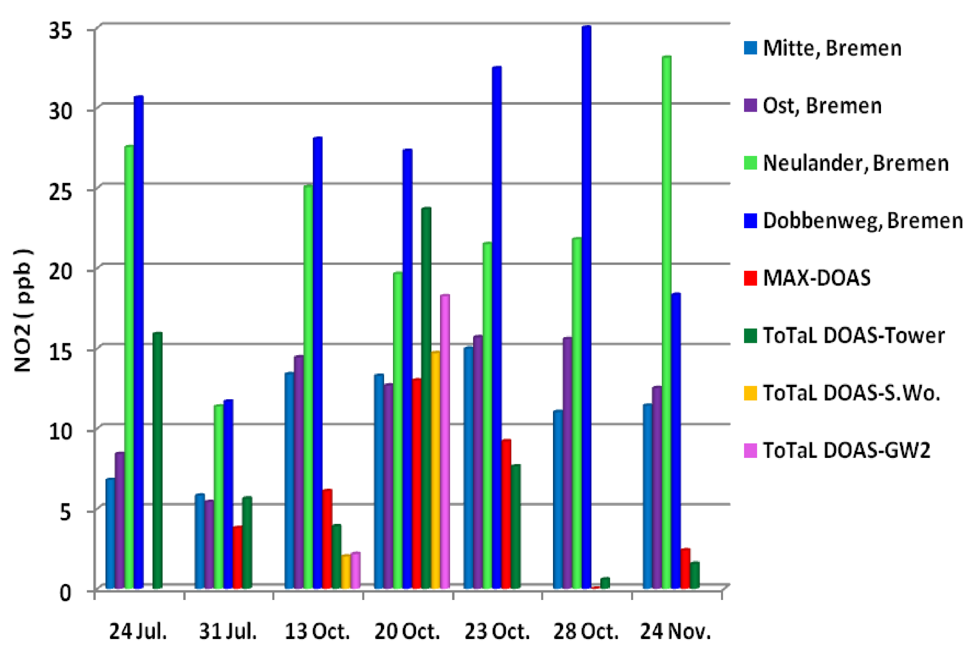
Figure (left): Geometry for the vertical scan of the drop tower. Figure (right): Retrieved concentrations of NO_2 for the scan on October 23, 2008 showing in average decreasing values for larger light paths (higher altitudes).



First Results and Comparison to Complementary Data

ToTaL-DOAS measurements of NO_2 were carried out on seven days between July and November 2008. The figure to the right shows as an example the diurnal variation for October 13. Circles indicate the different targets used on this day. While for measurements towards the Student Wohnheim and the GW 2 a small temporal variation was found, the target Drop Tower shows rapidly changing and significantly higher NO_2 values. Since this was a quite typical behaviour for the whole period a reasonable explanation is the changing traffic on the highway close to the tower.





The lower left figure shows the comparison between ToTaL-DOAS, MAX-DOAS and selected *in situ* stations located in the city centre of Bremen. The agreement between MAX-DOAS and ToTaL-DOAS is quite reasonable both in temporal variation and in absolute values. *In situ* measurements usually reveal much higher NO_2 values since there are closer to the sources (most of them are located a few meters next to the main streets in Bremen). Only for October 20, when there was a strong wind blowing from the city centre to the campus site, all different methods yield similar results. In order to better assess the accuracy of the method in the near future a new *in situ* instrument will be installed at the iup building.

Conclusions

• ToTaL-DOAS measurements of NO_2 are possible over very short distances, but restricted to sunny days.
• Comparison to complementary measurements of NO_2 show reasonable agreement.
• NO_2 concentrations show very high day-to-day variation
• Vertical scanning of a tower can be applied to retrieve the vertical profile of NO_2

Selected References

• Frins, E., U. Platt, and T. Wagner: High spatial resolution measurements of NO2 applying Topographic Target Light scattering Differential Optical Absorption Spectroscopy (ToTaL-DOAS), Atmos. Chem. Phys., 8, 7959-7601, 2008.
• Muhammad Umar Javed, ToTaL DOAS Measurements in Bremen, Master Thesis, University of Bremen, 2009.

Acknowledgement

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see also: www.iup.uni-bremen.de/doas