

Experimental imaging DOAS observations over Bremen

E. Peters¹, M. Ostendorf¹, A. C. Meier¹, A. Schönhardt¹, F. Wittrock¹, A. Richter¹, and J. P. Burrows¹

¹ Institute of Environmental Physics / Remote Sensing, University of Bremen, Otto-Hahn-Allee1, D-28359 Bremen, Germany

Email: Enno.Peters@iup.physik.uni-bremen.de



Introduction

Measurement principle: Differential Optical Absorption Spectroscopy (DOAS)

- Based on Lambert-Beer's law
- High-frequency part of (known) absorption structures σ are fitted to optical depth τ
- DOAS equation (I and I_0 are measured):

$$\tau_{\text{meas}} = \ln\left(\frac{I_0}{I}\right) = \sum_i \sigma_i(\lambda) \cdot SC_i + \text{polynomial} + \text{residual}$$

- Result: Slant columns $SC_i = \int \rho_i \cdot ds$ (absorber concentration ρ integrated over light path s)
- I_0 measured usually in zenith direction
- Current Multi-Axis (MAX-DOAS) instruments are able to point in any direction allowing several elevation and azimuth directions

Limitations of current MAX-DOAS instruments:

- Only one measurement in a certain pointing direction per time
- Full hemispheric coverage not possible as being too time-consuming
- Vertical scans (sequence of different elevations) performed in limited azimuthal directions only, or horizontal scan (sequence of different azimuths) performed in limited elevations

Aim of this work:

- Using an imaging spectrometer to perform measurements in multiple viewing directions simultaneously
- In addition: Mounting the entrance optics on a pan-tilt-head
- Full hemispheric coverage on the time scale of minutes

Instrument

Instrument characteristics:

- Adaptation from an air-borne DOAS instrument [1].
- Outdoor parts: Entrance optic (Camera objective, 48° FOV) mounted on commercial ENEO VPT-501 pan-tilt-head, 100°/s)
- Optical fiber bundle consisting of 38 single glass fibers vertically aligned in the same sequence at either end (35 mapped on CCD)
- allows optical imaging and flexible positioning of the instrument
- Indoor parts: Acton SP-2300i imaging spectrograph (temperature stabilized to 35°C, 413-499 nm, 0.7-1 nm resolution) in combination with a frame-transfer CCD (PhotonMAX) camera (cooled), electronics, computers.

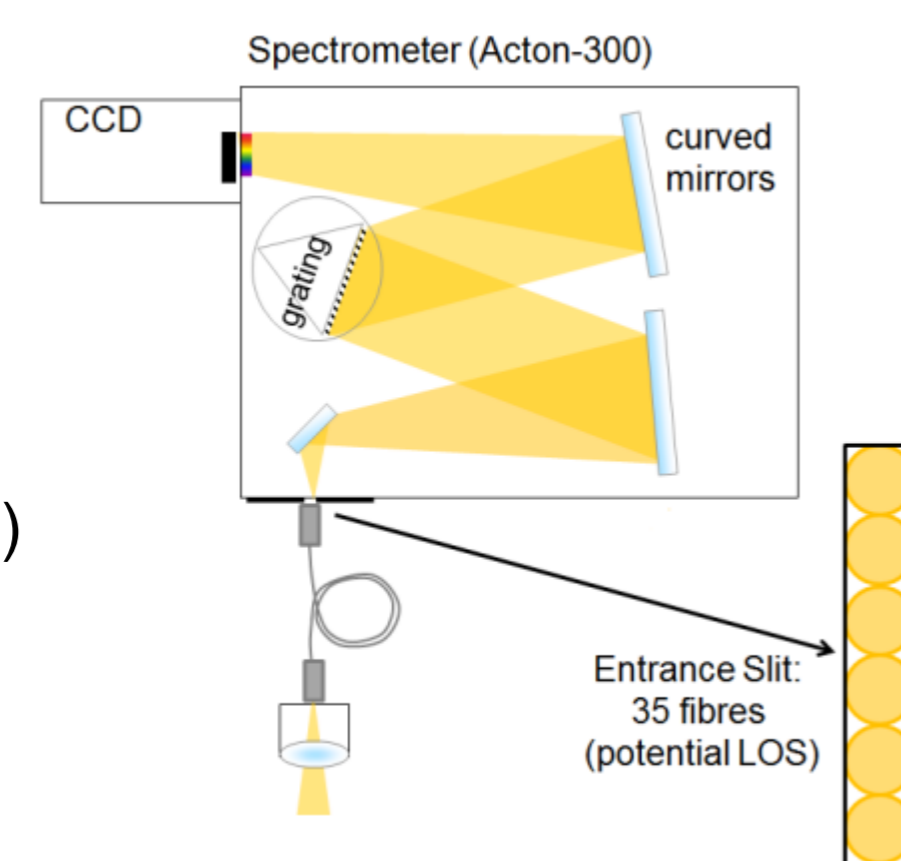


Fig 1: Sketch of the instrument comprising entrance optic (outdoor), optical fiber, and spectrometer (indoor) [3].

Advantage of the instrument:

- Due to the combination of special fiber bundle and imaging spectrometer the spatial information of the radiance is retained
- 35 equally spaced vertical viewing directions (elevation angles) of 1.2° each
- Pan-tilt-head allows azimuthal changes while 35 elevations are measured simultaneously
- Full hemispheric coverage each 6 minutes.
- Installed at roof of IUP-Bremen building from June to July 2014.



Fig 2: Entrance optic providing 35 elevations simultaneously mounted on pan-tilt-head for azimuthal movement.

Azimuthal NO₂ distribution over Bremen

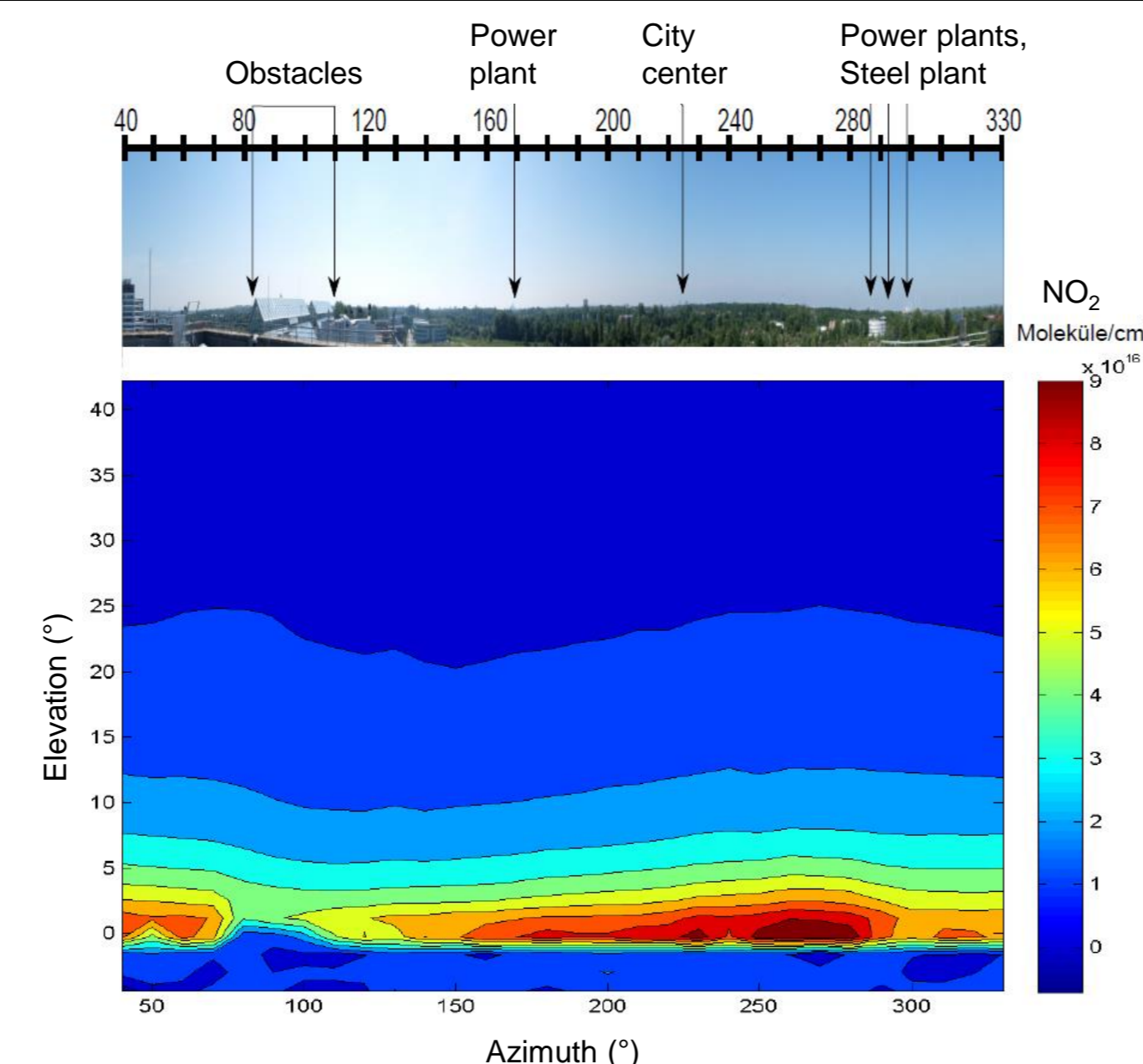


Fig 3: Top: Panorama view from IUPB roof. Bottom: Mean NO₂ SCs on 23 July 2014 [2]. (Negative elevations obstructed by building)

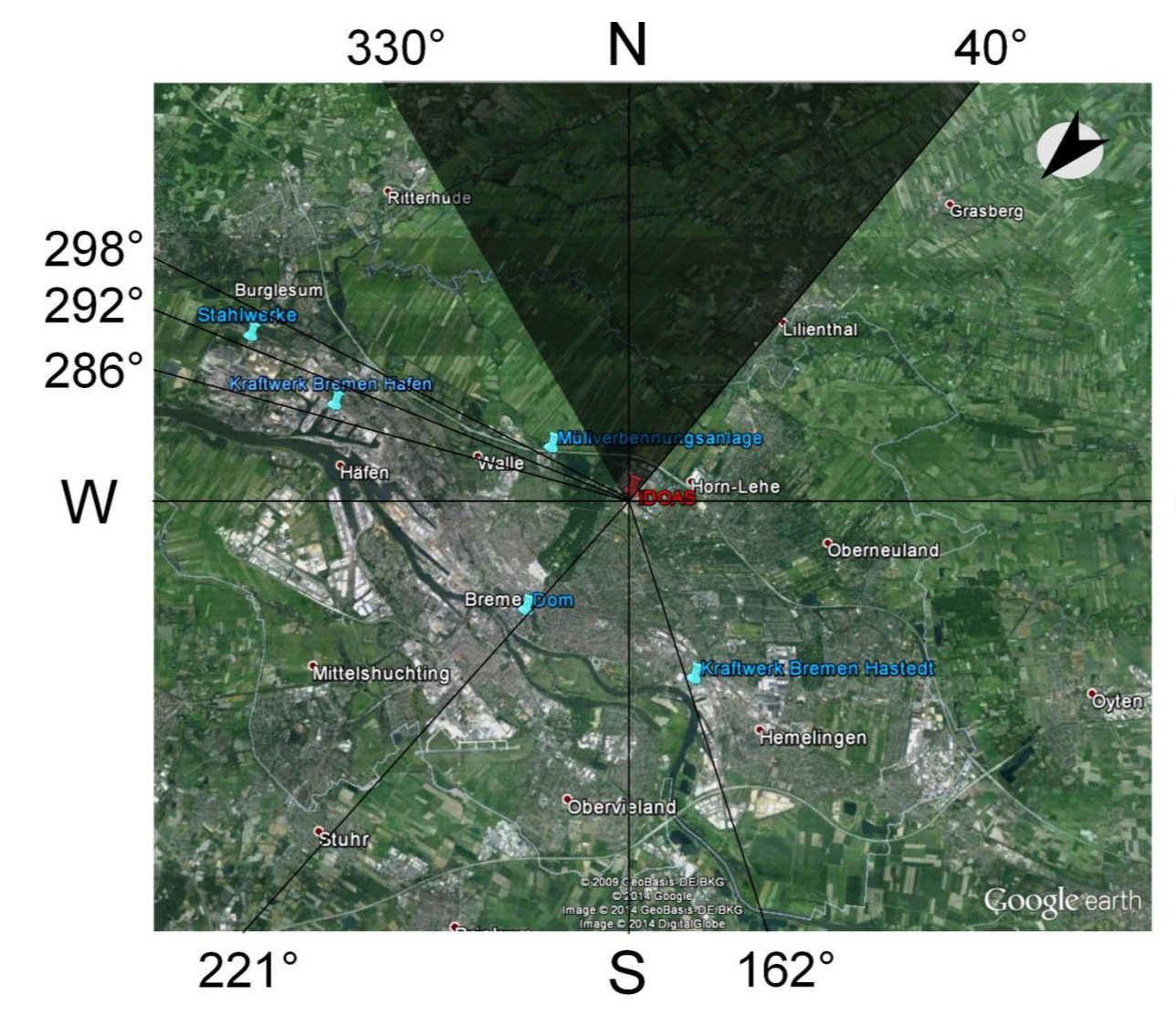


Fig 4: Aerial view of Bremen. Azimuthal viewing directions and possible point sources are indicated [2].

- Azimuthal scans from 40° N to 330° N in 10° steps, integration time: 10 s
- No measurements between 330° and 40° N due to obstruction by building
- Pan-tilt-head allows recording zenith spectra after each azimuthal scan
- Using closest zenith spectrum as reference I_0
- NO₂ DOAS fits: 425-450 nm, resp. 425-490 nm (O₃, NO₂, O₄, H₂O, Ring, intensity offset)

- Wind direction at 23 July 2014 (Fig. 3 and 5): North-East (as indicated in Fig. 4)
- Generally larger NO₂ in direction of the city
- Largest NO₂ pollution from near-by power and steel plants (in Fig. 3 shifted because of wind direction)
- No enhanced NO₂ detected for power plant further away (see Fig. 4)
- Azimuthal distribution of other trace gases (O₄, H₂O) demonstrate influence of measurement geometry (position of the sun)

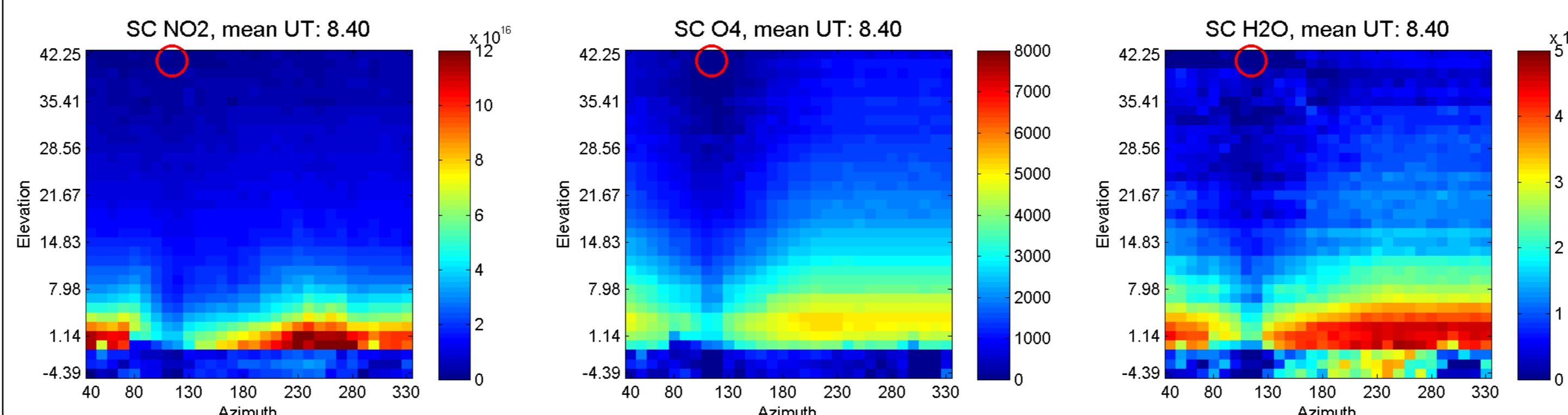


Fig 5: Example of a single 6 min azimuthal scan from 23 July 2014 (morning). Left: NO₂, middle: O₄, right: H₂O. Cloud-free sky. The position of the sun is indicated by a red circle. (DOAS fit here: 425-490 nm).

References

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- [2] Ostendorf, M.: Azimuthal monitoring of trace gases in the atmosphere using an imaging DOAS instrument in Bremen, Bachelor thesis, University of Bremen, 2014.
- [3] Altube, P., Aircraft measurements of tropospheric NO₂ with an imaging DOAS instrument, Master thesis, University of Bremen, 2012.
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Intercalibration & Outlook

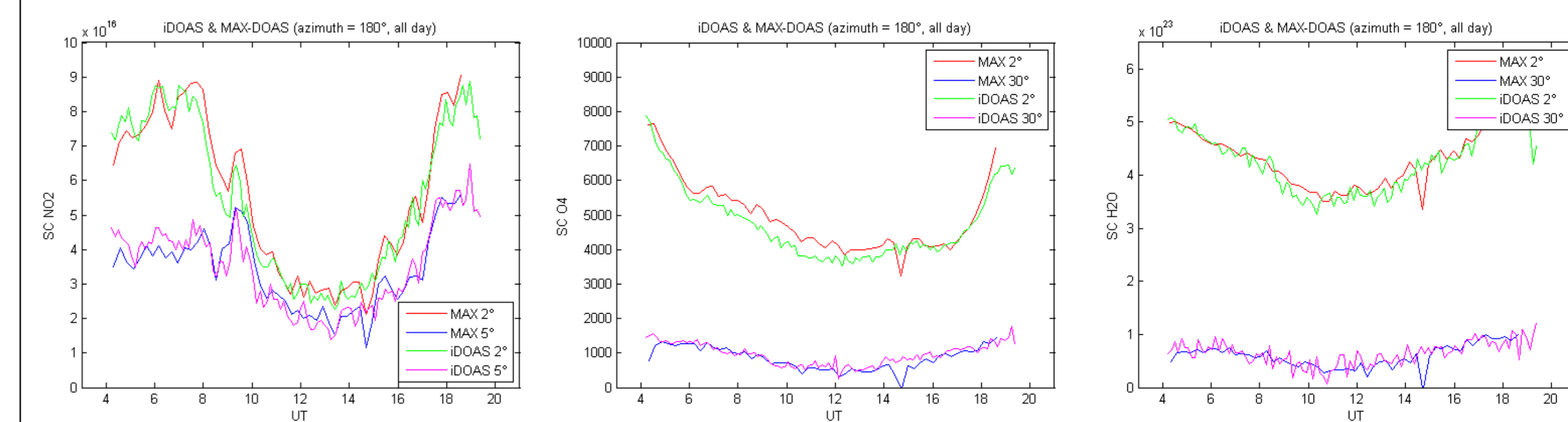


Fig 6: Intercalibration of the resulting slant columns of NO₂ (left), O₄ (middle) and H₂O (right) in 180° azimuth and different elevation angles between the imaging DOAS instrument and a MAX-DOAS instrument operated close-by.

Intercalibration results:

- Intercalibration was possible with a close-by MAX-DOAS instrument operated routinely
- Generally good agreement between MAX-DOAS and experimental imaging DOAS instrument was found for all trace gases
- MAX-DOAS and imaging DOAS did not point at exactly the same time in the same direction
- MAX-DOAS spectra were recorded using full vertical (software) binning of the CCD rows (expected to yield better results due to averaging)

Possible further applications:

- Measurements of O₄ could be used to study the effect of viewing geometry and aerosols on the resulting slant columns
- Retrieving aerosol information simulating O₄ via radiative transfer model (here: SCIATRAN)
- Possibly cloud flags could be elaborated when investigating cloudy days

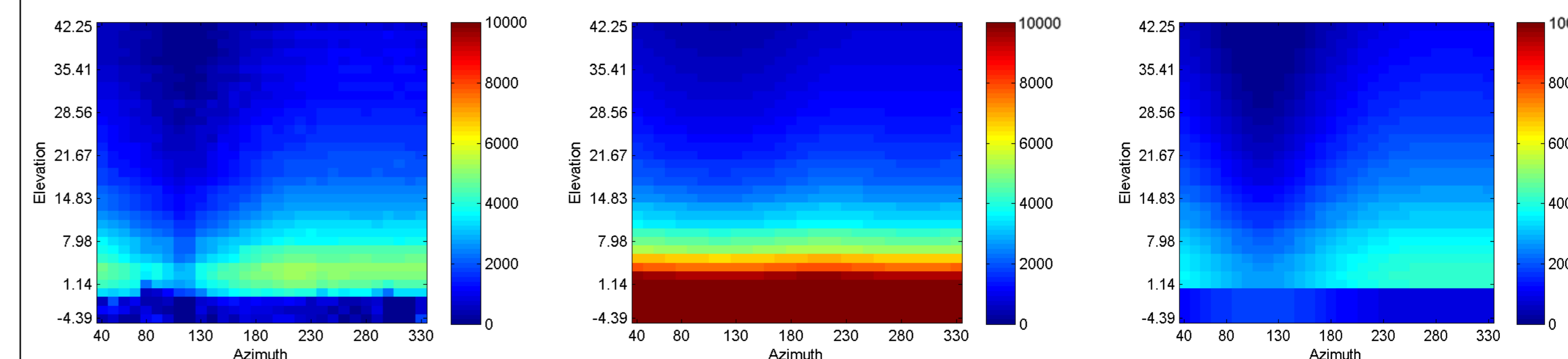


Fig 7: Left: Measured O₄ slant columns at 23 July 2014, 8.40 UT. Middle: Simulated O₄ slant columns for Rayleigh atmosphere and same viewing conditions (only positive elevations). Right: Same simulations including aerosols.

Conclusions

- Good vertical and horizontal coverage achieved at high speed (6 minutes) overcoming the limitation of current ground-based MAX-DOAS instruments
- Full hemispheric detection, i.e. vertical as well as azimuthal distribution, of tropospheric NO₂ over Bremen possible, identification of emission sources
- Good agreement found for coinciding viewing directions with close-by MAX-DOAS instrument
- Temporal evolution of NO₂ pollution can be monitored (duration of complete hemispheric scan ca. 6 minutes → much faster than NO₂ lifetime)
- Demonstrating effects of viewing geometry on retrieved slant columns in different directions
- Outlook: Comparing measured and simulated O₄ → adjusting (i.e. retrieving) aerosol scenario

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