Spatial distributions of NO₂ in emission plumes observed by imaging DOAS from aircraft

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Motivation

Objectives

- Measurements of tropospheric trace gases, e.g. NO₂, at good spatial resolution and coverage
- NO₂ pollution mapping, identification of source regions and source strengths
- Detailed investigation of spatial variability of NO₂ column amounts

Advantages of aircraft measurements and the IUP imaging DOAS instrument AirMAP

- High spatial resolution ~100 m (down to <30 m) at useful spatial coverage
- Many viewing directions observed at the same time within a broad stripe below the aircraft

53.7

-atitude 53.6

53.5

53.7

Latitude 9:85

SE, 120°

7.9

53.5 11-12 kn

Full coverage with no data gaps independent of flight altitude

NO₂ observations downwind of a power plant

Power Plant Location:

Wilhelmshaven 53.565°N, 8.147°E

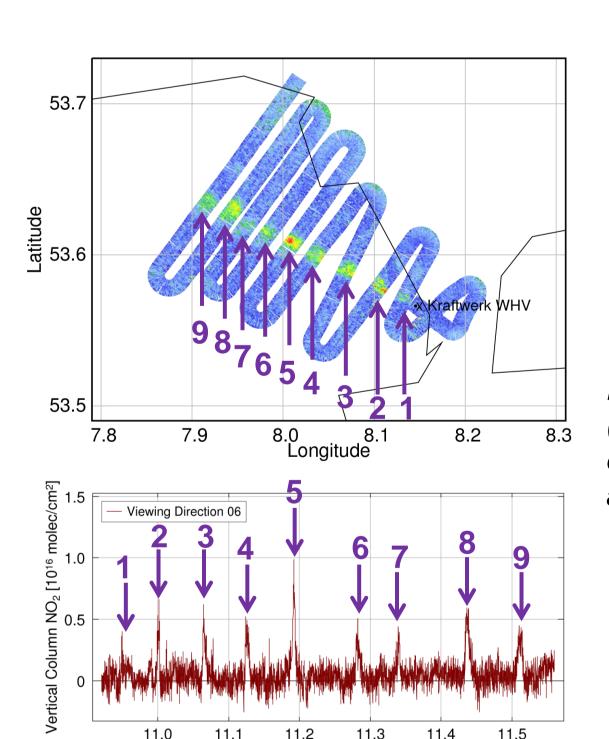
Emission report (http://prtr.ec.europa.eu): Emission of NO_x/NO_2 : 1.900-2.650 t/a NO is emitted from the power plant and is subsequently converted to NO₂

Observations of the NO₂ emission plume Flight on 24.08.2013

Flight pattern #1: along the plume and back Flight pattern #2: crossing the plume several times at different distances from the stack

Spatial distribution of NO₂

- NO₂ enhancement downwind of the power plant stack clearly visible
- Localised NO₂ vertical column maxima reach up to 1.10¹⁶ molec/cm²
- Distribution is strongly inhomogeneous
- The same localised NO₂ maximum is probably observed twice in Pattern #1
- The plume evolution differs strongly from uniform Gaussian plume dispersion



VC NO₂ [10¹⁵ molec cm⁻²] Figure (above): Spatial distribution of NO2 vertical columns downwind of the Wilhelmshaven power plant on 24.08.2013 for two flight patterns, #1 along the plume direction (top) and #2 crossing the plume (bottom) at different distances. The arrows mark the flight direction.

8.0

Longitude

NO₂ VC on 24.08.2013

Flight

pattern #1

Kraftwerk WHV

Flight

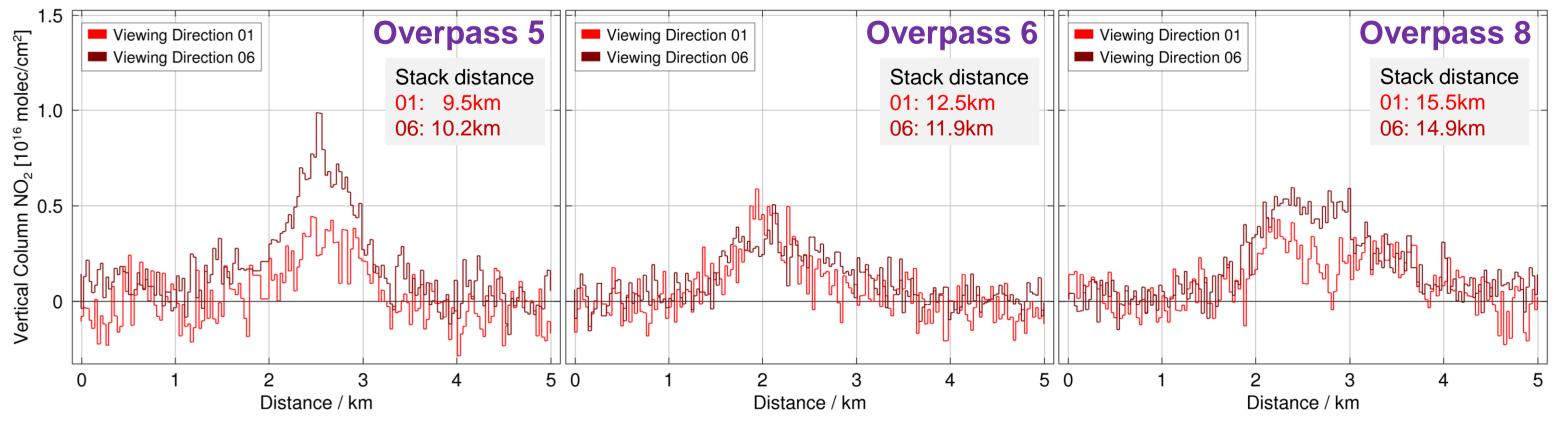
8.2

pattern #2

8.3

Figure (left): Numbered overpasses in flight pattern #2 (top) and time series of NO₂ vertical columns for example viewing direction 06 showing maxima in NO₂ amounts for the 9 individual overpasses.

> Figure (below): Plume cross sections of the NO2 vertical column amount observed during flight pattern #2 for two different viewing directions, 01 and 06, at three different overpass locations, overpass 5 (left), overpass 6 (middle) and overpass 8 (right). The distance given on the horizontal axis is the track length along flight direction, i.e. across the plume, with individual zero points for each overpass.



- Large differences in integral NO₂ amounts are observed between the viewing directions, i.e. for only slightly different distances from the exhaust stack (see insets in figures)
- With increasing distance from the stack (overpass 5 to 8), the plume slightly broadens
- Overpass 6 shows much less NO₂ than overpass 5, although further away from the stack, while generally, conversion from NO to NO₂ leads to an increase of NO₂ with time and distance

Universität Bremen

The AirMAP instrument in the Cessna aircraft

AirMAP: Airborne imaging DOAS Measurements of Atmospheric Pollution

Instrument developed at IUP Bremen in 2011

Flight campaigns in June 2011 (AWI Polar-5 aircraft) and August 2013 (FU Berlin Cessna aircraft)

Cessna 207 Turbo (D-EAFU)

Speed: typ. 50-60 m/s

Ceiling height: 6000m Operating height: typically 800 – 1500 m (during the AirMAP campaign 2013)

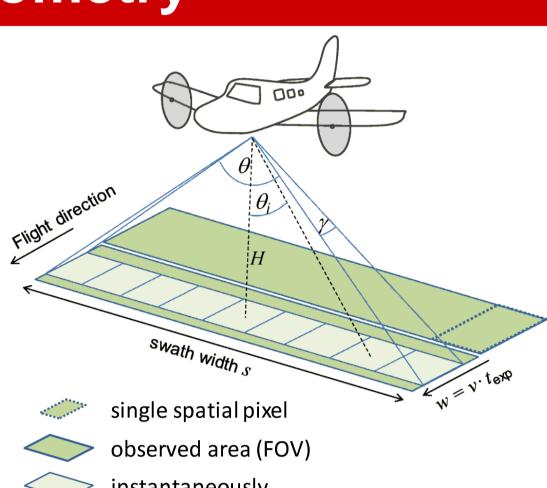
Owner & Operator: FU Berlin since 1988

Photograph*: The Cessna aircraft at Flugplatz Schönhagen Brandenburg, Germany. *by Mareike Ostendorf



Instrumental setup and viewing geometry

- Optics: Wide angle objective and fibre bundle (35 fibres)
- 2 nadir ports: spectrometer objective and picture camera
- Acton 300i imaging spectrometer
- Spectral window: 412 453nm; 0.5-1.0nm resolution
- Detector: Frame transfer (FT), 512x512 pixel, 8.2x8.2 mm²
- Field of view: ~48° across track (θ), ~1.5° along track (γ)
- Swath width: on the order of flight altitude H
- Viewing directions: max. 35 LOS (line of sight)
- Averaging across track: combining fibres to 9 LOS (θ_i)
- Exposure time t_{exp}: 0.5 s
- Flight speed typ. 60 m/s
- Spatial resolution: <100m across track (at ~1km flight altitude, 9 viewing directions), ~ 30 m along track
- Positioning information: from GPS sensor and gyrometer to determine correct geolocation



instantaneously observed area (iFOV) Figure:

Sketch of the viewing geometry

The AirMAP instrument allows gap-free measurements along and across flight direction

NO₂ retrieval

Retrieval Settings

Fitting window: 425 - 450 nm

Trace gases: NO₂ (293K), O₃ (241K), O₄ (296K), H₂O (HITRAN)

Atmospheric effects: Ring (SCIATRAN calculated), quadratic polynomial, intensity offset

Reference I₀: rural scene from same LOS Slit function: individual for each LOS

Detection Limit for NO₂

Slant Column detection limit ~10¹⁵ molec/cm²; optical density rms on the order of 10⁻³

Air mass factors, AMF (SCIATRAN)

Rayleigh atmosphere, 1 km NO₂ box profile, 5% albedo, SZA and LOS dependence.

Emission estimates

NO₂ emission flux calculations

- Flux calculations at different distances from stack
- Approximation of source strength is achieved via discrete sum over the product of vertical columns VC, wind speed u and path length dl.

$$Q \cong \int_{L} VC \cdot \vec{u} \cdot d\vec{l} \approx \sum_{i} VC_{i} \cdot \vec{u} \cdot d\vec{l}_{i}$$

Example calculation for overpass 5

- 9 different values for Q from 9 viewing directions, i.e. different distances from stack the stack (pattern #2)
- Calculated fluxes vary between 1.8 and 5.5·10²³ molec/s.

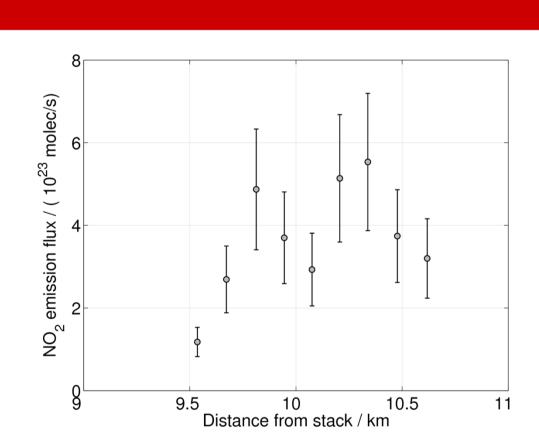


Figure: NO₂ emission flux calculated for different distances from the exhaust stack within overpass 5. The emission results are strongly variable.

Summary and Outlook

- NO₂ vertical column amounts have been observed from aircraft downwind of a power plant.
- Imaging capabilities of AirMAP allow plume observations at good spatial coverage and resolution.
- The spatial NO₂ distribution is non-uniform and varies strongly along the plume.
- With increasing distance from the stack, the plume slightly broadens.
- Instead of gradually increasing, the NO₂ is often confined in bubble-like structures.
- The results have implications for the importance of emission sources and downwind chemistry, because localised amounts of NO₂ lead to different effects than a smoothly averaged distribution.
- Possible reasons for the non-uniform distributions and plume evolution include source variability, chemical transformations and local meteorology.
- Further analysis of the plume structure will be performed including dynamics and plume chemistry.

Selected References

- P. Wang, et al: Measurements of tropospheric NO₂ with an airborne multi-axis DOAS instrument, Atmos. Chem. Phys., 5, 337-343, 2005.
- K.-P. Heue, et al: Direct observation of two dimensional trace gas distributions with
- an airborne Imaging DOAS instrument, Atmos. Chem. Phys., 8, 6707-6717, 2008.
- C. Popp et al.: High-resolution NO₂ remote sensing from the Airborne Prism EXperiment (APEX) imaging spectrometer, Atmos. Meas. Tech., 5, 2211-2225, 2012.
- A. Schönhardt et al: A wide field-of-view imaging DOAS instrument for continuous trace gas mapping from aircraft, accepted for Atmos. Meas. Techn. Disc., 2014.

Acknowledgements

The authors gratefully acknowledge financial support by the University of Bremen (Zukunftskonzept M8 PostDoc Projekt, Exzellenzinitiative des Bundes und der Länder) as well as by the BSH (Bundesamt für Seeschifffahrt und Hydrographie, Hamburg) MeSMarT project. Campaign support at Flugplatz Schönhagen and by Martin Gehrmann at AWI Bremerhaven, is gratefully acknowledged.