Aircraft imaging DOAS measurements of anthropogenic nitrogen dioxide

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1. Introduction

Objectives of aircraft imaging DOAS measurements:

- Retrieval of tropospheric trace gases, here nitrogen dioxide NO_2
- Applying Differential Optical Absorption Spectroscopy (DOAS) technique
- Mapping of NO_2 pollution sources, identification of source regions and strengths
- Satellite data validation, investigation of sub-pixel variability

Positive aspects of aircraft measurements and imaging DOAS

- High spatial resolution ~100 m (down to ~30 m) at useful spatial coverage
- Several viewing directions across track are observed simultaneously
- No data gaps occur along track

The iDOAS instrument in the Polar-5 aircraft

Aircraft Type:	Basler BT-67 / DC3
Length/Height/Span:	21 m / 5.2 m / 29 m
Speed & Altitude:	50-105 m/s; 100-19000 ft
Owner & Operator:	AWI, Germany; Kenn Borek Air Ltd. Canada

Photographs: (top) iDOAS installed in Polar-5 aircraft (bottom) Polar-5 in the hangar at Bremerhaven regional airport

2. Instrumental setup and viewing geometry

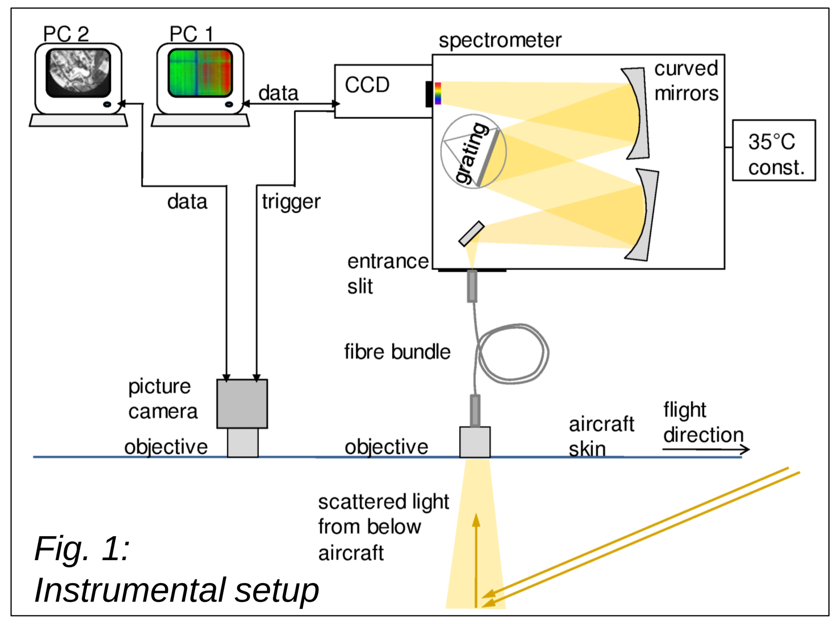


Fig. 1: Instrumental setup

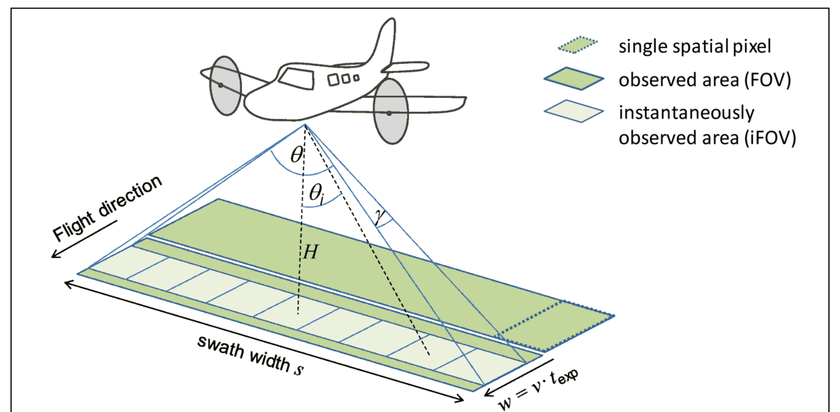


Fig. 2: The iDOAS viewing geometry

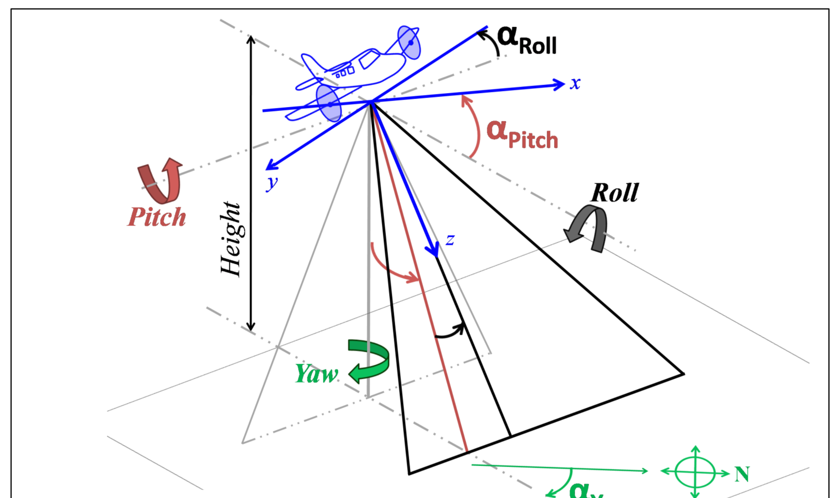


Fig. 3: How the aircraft angles affect the ground pixel location

Technical information

- Wide angle objective and fibre bundle (35 fibres) as entrance optics
- Acton 300i imaging spectrometer
- Grating 600l/mm, blazed @500nm
- Spectral window 415 - 455nm
- Spectral resolution 0.7 - 1.0nm
- Frame transfer (FT) CCD Detector, 512x512 pixels, 8.2x8.2 mm²

→Gap-free measurements (due to FT CCD) and flexible positioning in aircraft (due to sorted fibre bundle)

Viewing geometry

- 2 nadir ports: spectrometer &camera
- Geolocation: from GPS & gyrometer
- Viewing directions: max. 35 (typ. 9) lines of sight, (LOS, θ_i) from 35 fibres
- Field of view: ~48° across track (θ)
- Swath width: ~order of flight altitude H
- Exposure time t_{exp}: typ. 0.5s
- Spatial resolution: ~100 m and less

Computation of ground pixel location

- Consideration of the aircraft angles (pitch, roll and yaw) is required in addition to GPS position for correct determination of the geolocation
- Displacements of the ground pixel due to aircraft motions can be significant

3. NO_2 vertical columns and emission flux calculations above a power plant

NO_2 retrieval above a power plant

- Black coal power plant (848 MW) at Ibbenbüren, Germany (52°17'N, 7°45'E)
- Slant columns of NO_2 retrieved by Differential Optical Absorption Spectroscopy
- Large variability of NO_2 amounts across and along track is observed
- The NO_2 in the exhaust plume downwind of the power plant is clearly visible
- Transects through the plume are used for emission flux estimations

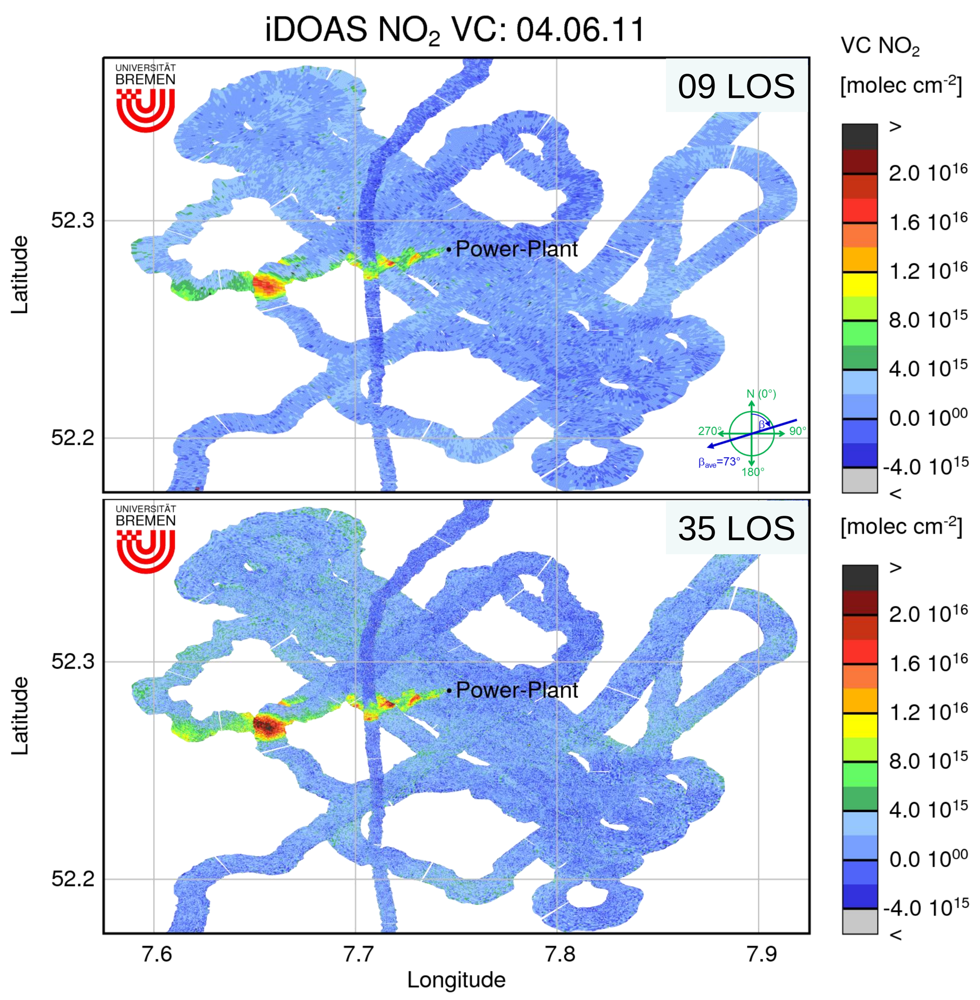


Fig. 4: NO_2 vertical column amounts along the flight track retrieved from the flight on 04.06.2011. Downwind from the power plant of Ibbenbüren, strong enhancement of NO_2 is visible. Average wind direction was about East North-East, see inset. Enhanced NO_2 is on the order of 10^{16} molec/cm² with maxima > $2\cdot10^{16}$ molec/cm². Top: Division of the field of view into 9 lines of sight (LOS) allowing spatial resolution of ~100m. Bottom: Consistent result for full spatial resolution of 35 LOS with ground pixel side length on the order of around 30m. Fine spatial variability of NO_2 amounts is resolved.

Retrieval Settings

Fitting window: 425 – 450 nm

Trace gases:
NO_2 (293K), O_3 (241K), O_4 (296K), H_2O (HITRAN)

Atmospheric effects:
Ring (SCIATRAN calculated), intensity offset

Polynomial: quadratic

Reference I_0: rural scene from same LOS

Slit function: individual for each LOS

Detection Limit for NO_2

Slant Column (SC) detection limit: ~10^{15} molec/cm²
Optical density RMS: on the order of 10^{-3} for a single measurement of 0.5s and an individual LOS.

Air mass factors, AMF (SCIATRAN calculations)

Rayleigh atmosphere, 1 km NO_2 box profile, 5% albedo, SZA and LOS dependence.

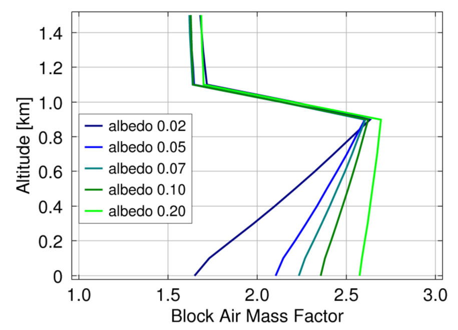
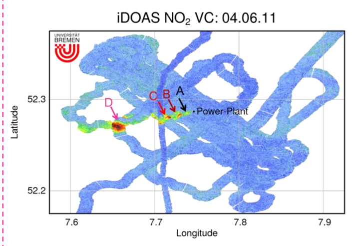


Fig.5: Block AMF for different albedos at 40° SZA and 1.1km flight altitude. AMF differences between box profile and elevated Gaussian plume are ~10%.

Transect positions through plume at different distances from the stack around 10:00 UT used for emission flux calculations:



NO_2 emission flux calculations

- Based on Gaussian plume dispersion model
- Mean wind speed & direction determined using COSMO-DE model wind data and weighting by NO_2 profile (Gaussian shape, cp. Fig.6)
- Flux calculations performed at different distances from stack

$$c(x,y,z) = \frac{Q}{2\pi\sigma_y\sigma_z u}\exp\left(-\frac{y^2}{2\sigma_y^2}\right)\exp\left(-\frac{z^2}{2\sigma_z^2}\right)$$

Eq. 1: Gaussian distribution of concentration c

Dispersion of concentration c across plume (y) and over altitude (z) is taken into account, with source strength Q, wind speed u and spread σ_y and σ_z. Along the wind direction x only advection is considered.

$$Q \cong \int_L VC\cdot\vec{u}\cdot d\vec{l} \approx \sum_i VC_i\cdot\vec{u}\cdot d\vec{l}_i$$

Eq. 2: Derived using Gaussian divergence theorem

Approximation of source strength is achieved via discrete sum over product of vertical columns (VC), wind speed and path length dl.

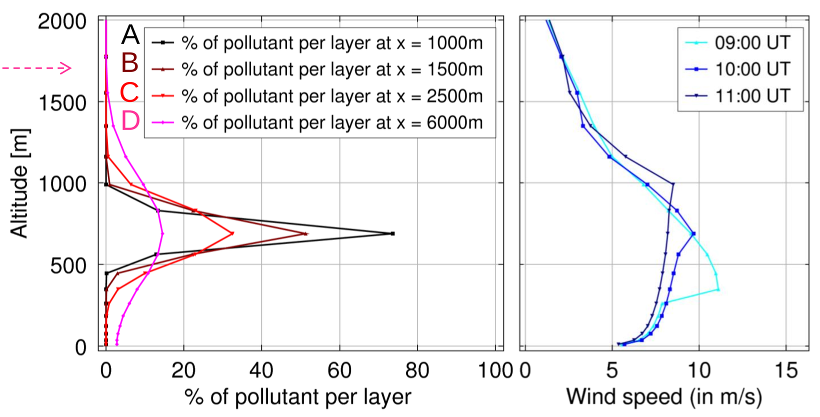


Fig. 6: Relative NO_2 altitude distribution inside the plume at four different distances from the stack (left) and wind speeds from the COSMO-DE model (right). NO_2 profiles are used as weighting factors to determine mean wind speed and direction.

→ Estimated NO_2 emissions: Q_{NO2} ≈ 2100-2400 T/a
→ Emissions of NO_x (using NO/NO_2 ≈ ¼) : Q_{NOx} ≈ 2600-3000 T/a
→ Results are in agreement with E-PRTR[#]

[#]European Pollutant Release and Transfer Register

4. NO_2 above inhabited and rural areas

NO_2 above Hamburg and Northern Germany

- Urban NO_2 SC maxima lie around $1\cdot10^{16}$ molec/cm²
- Enhanced NO_2 above Hamburg and close to the airport
- Strong spatial variability of NO_2 is observed

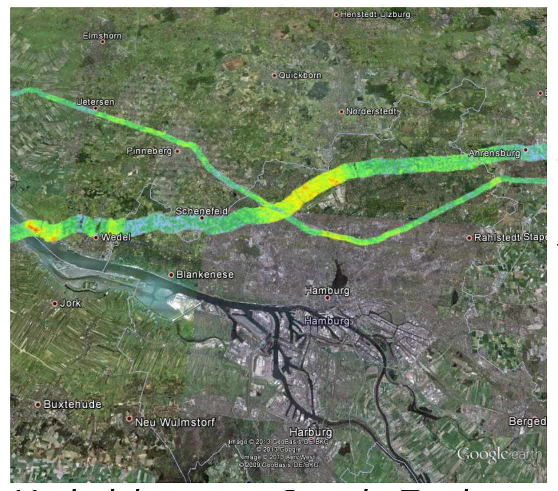


Fig.7 (left): NO_2 observations during two overflights over the city of Hamburg (same colour scale as Figs. 4 & 9). The flight altitude determines the width of the swath.

Underlying map: Google Earth

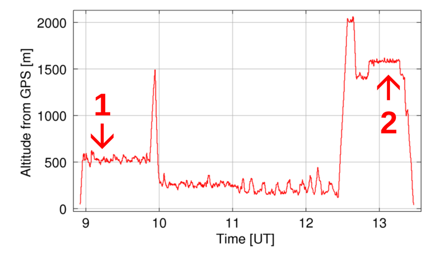


Fig.8: Flight altitude on 09.06.2011

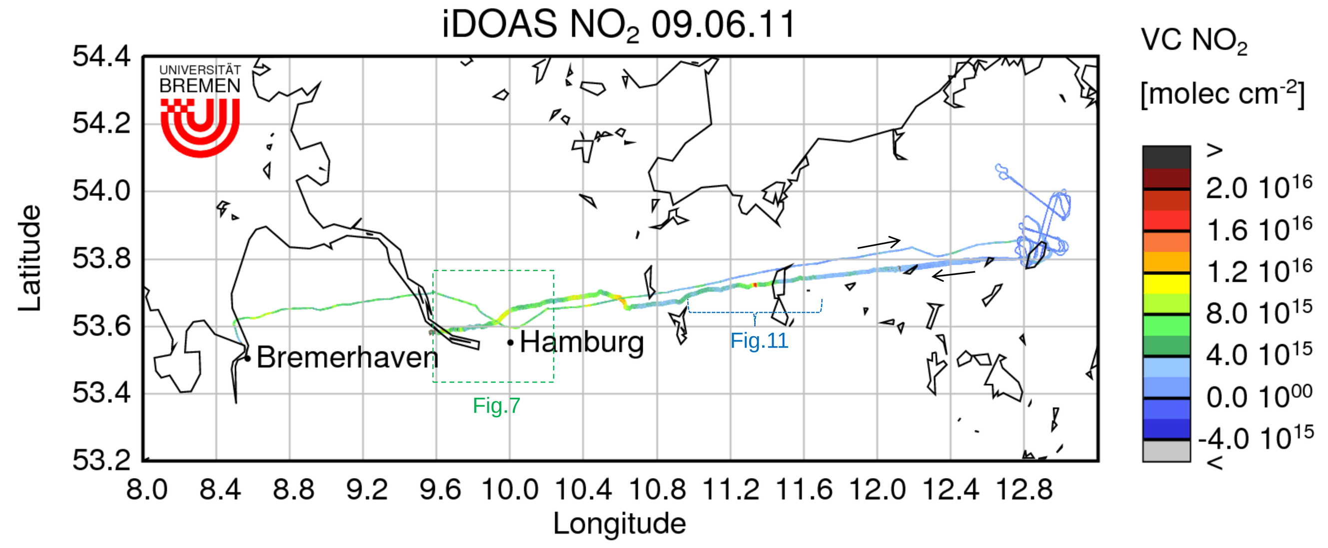
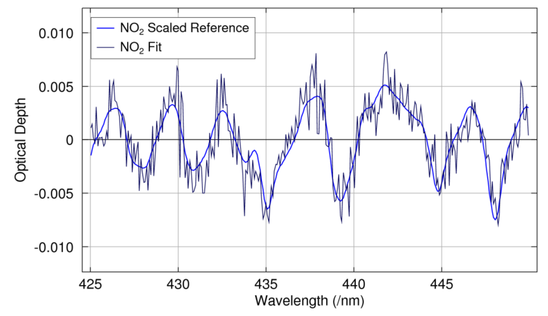


Fig.9: NO_2 vertical columns observed on 09.06.2011. Strong differences in NO_2 results are seen: much smaller amounts above rural areas, e.g. for the East part of the flight track, than closer to cities, e.g. around the Hamburg area. Not all NO_2 enhancements can be directly assigned to local sources, also transported NO_2 is observed. Green box: Region shown in Fig.7. Blue bracket: Section shown in Fig. 11 with confined NO_2 enhancement.

Fig.10: Example NO_2 retrieval result from the central LOS, on 09.06.2011 at 12:49:23 UT. NO_2 SC = $3.8\cdot10^{16}$ molec/cm², Fit error 3.8%, RMS=$1.77\cdot10^{-3}$.



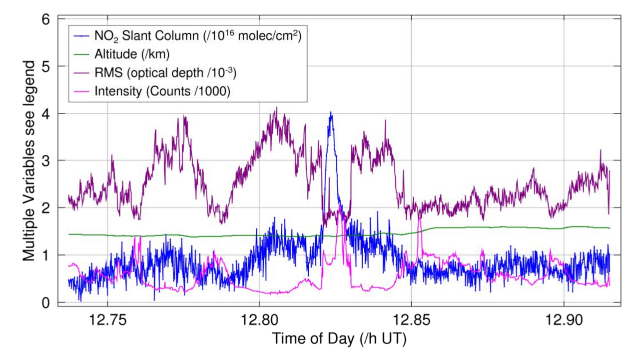


Fig.11: NO_2 columns (blue), altitude (green), RMS (purple) and intensity (pink) on 09.06.2011 around 12:50UT.

NO_2 enhancement at 12.82UT is partly situated within a cloud (amplified intensity) with good fit quality (reduced RMS) as expected for bright scenes. The NO_2 amount is much larger than above/in other clouds nearby. Radiative transfer calculations will separate path length effects from actual NO_2 increase.

5. Summary & Outlook

Summary

- Imaging DOAS instrument shows good imaging quality and good performance for NO_2 measurements
- Aircraft pitch, roll and yaw angles are fully taken into account for correct ground geolocation
- NO_2 column amounts have been retrieved, pollution sources are observed (power plant, cities, etc)
- Further findings: Large spatial NO_2 variability, consistent NO_2 retrieval results for different LOS divisions, transported NO_2 within a cloud away from local sources, consistently low NO_2 above rural areas
- NO_2 emission fluxes are calculated for a power plant point source in agreement with emission reports

Activities for the future

- Air mass factor consideration will be refined in future analyses
- Further dedicated campaigns will be conducted with the imaging DOAS instrument above pollution sources

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