Formaldehyde and nitrogen dioxide over the western Pacific: SCIAMACHY and GOME-2 validation

E. Peters[1], F. Wittrock[1], K. Großmann[2], U. Frieß[2], A. Richter[1], J. P. Burrows[1]

[1] Institute of Environmental Physics/Remote Sensing, University of Bremen, Otto-Hahn-Allee1, D-28359 Bremen, Germany
[2] Institute of Environmental Physics, University of Heidelberg, Im Neuenheimer Feld 229, D-69120 Heidelberg, Germany

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

The TransBrom campaign

Campaign facts:

- Ship-borne campaign onboard the research vessel „Sonne“
- Accross western Pacific from Tomakomai (Japan) to Townsville (Australia)
- 9 - 24 October 2009
- Organized by GEOMAR Kiel (focusing on short-lived bromine compounds and flux ocean – stratosphere)

IUP-Bremen MAX-DOAS instrument:

- 2-channel instrument, Y-shaped optical fibre bundle leads light collected by telescope unit into two spectrometers
- Visible: 400-573 nm, 0.8 nm resolution (retrieved species: NO_2)
- UV: 315-384 nm, 0.4 nm resolution (retrieved species: HCHO)
- Telescope unit installed at monkey deck, pointing portside (to the east for most of the cruise)

Fig. 2: MAX-DOAS telescope unit

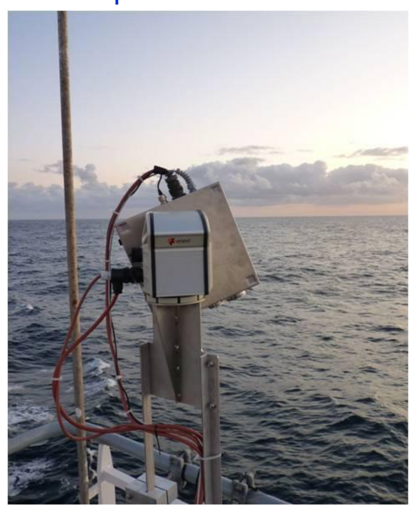


Fig 3: Cruise track

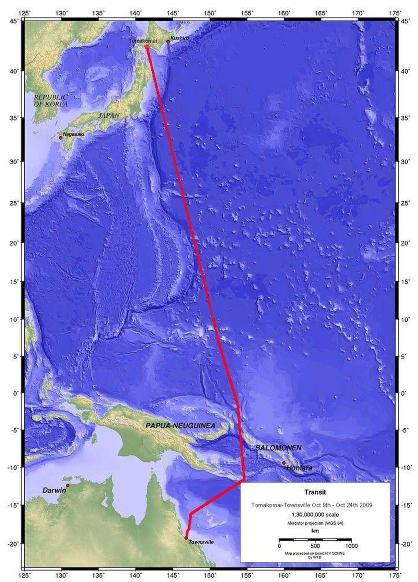


Fig 1: RV Sonne

Presented here:

- MAX-DOAS measurements of NO_2 (stratospheric and tropospheric) and HCHO
- Results of profile retrieval (optimal estimation) for tropospheric NO_2 and HCHO → VCs and profiles
- Estimation of background levels
- Validation of corresponding GOME-2 and SCIAMACHY measurements

Validation I – Stratospheric NO_2

MAX-DOAS measurements:

- Two stratospheric NO_2 columns per day → a.m. and p.m. values (highest sensitivity during twilight, 88° < SZA < 92°)
- Evening value larger than morning value because of slow N_2O_5 photolysis → Case study 15 Oct: Linear NO_2 increase of ≈ 8.7 x 10^{13} molec/cm²/h

GOME-2, SCIAMACHY validation:

- All pixels averaged within 150 km around ship's position at overpass time
- Same latitudinal variation as MAX-DOAS values
- Differing by 1-1.7% from each other
- On average marginally lower than MAX-DOAS a.m. values due to satellite overpass time and NO_2 diurnal cycle

Fig. 4: Cruise track and GOME-2 pixels around ship's location (r<150 km)

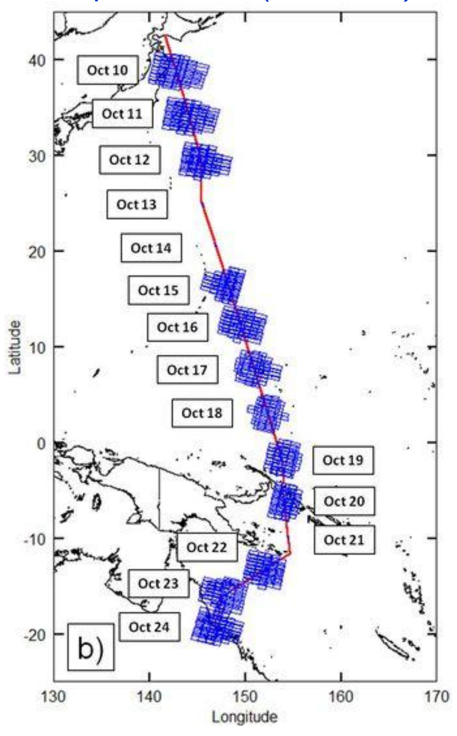


Fig. 5: Stratospheric NO_2 VCs from MAX-DOAS (am: magenta, pm: blue), GOME-2 (green) and SCIAMACHY (red)

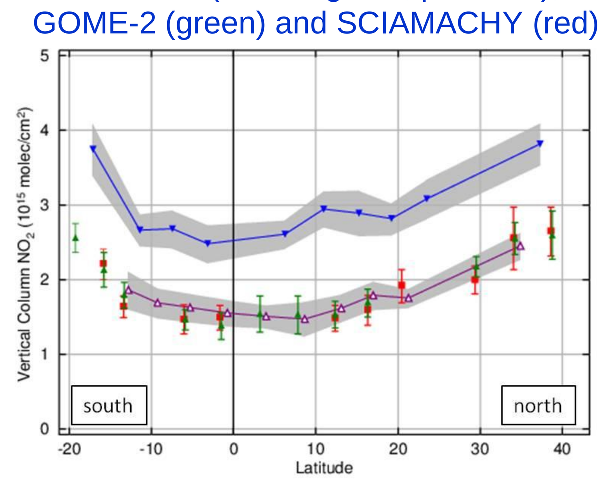
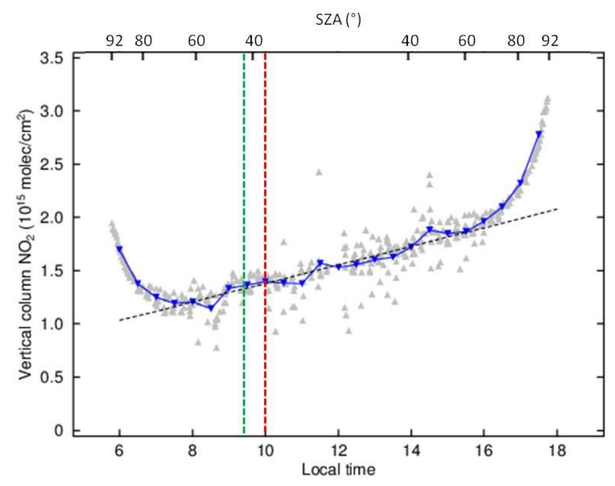


Fig. 6: Case study: Oct 15 all day NO_2 VCs (red and green lines = overflight times)



Validation II – Tropospheric NO_2

Fig. 7: MAX-DOAS NO_2 slant columns in 2°, 8°, 30° elevation angles (red arrows = passing of other ships, dashed line = detection limit)

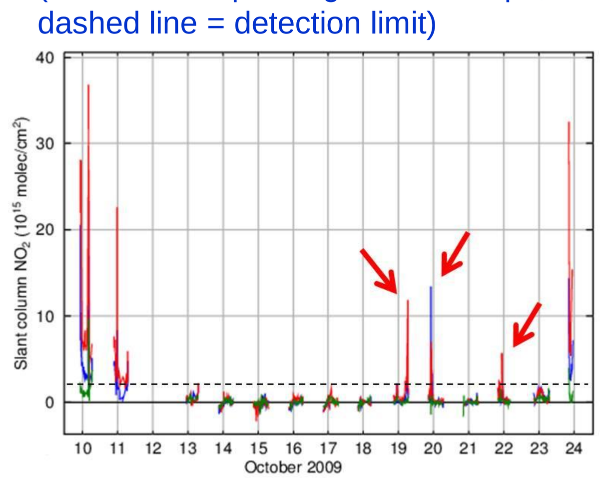
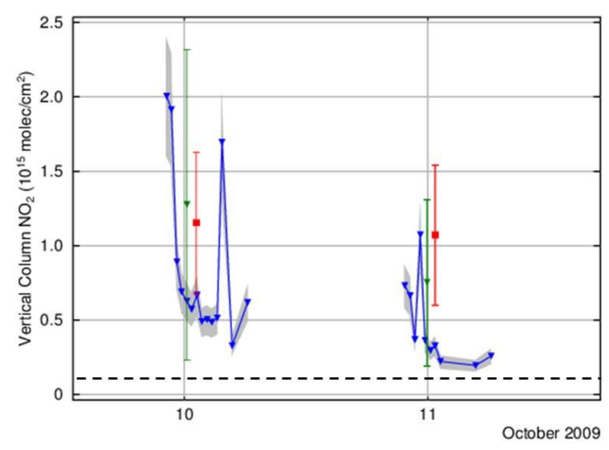


Fig. 8: NO_2 vertical columns for Oct 10-11 (MAX-DOAS = blue, GOME-2 = green, SCIAMACHY, red)



MAX-DOAS slant columns:

- Higher values only in the beginning and end (influence from land)
- Below detection limit (DL) over the open ocean
  → Exception: 3 events, passing of other ships
  → Estimated oceanic background concentration from DL
  VC = 1.3 x 10^{14} molec/cm²
  corresponding to VMR ≈ 50pptv (for 1km block profile)
- Estimated background agrees with with GOME-2 monthly average

Tropospheric vertical columns (VCs):

- VCs, profiles retrieved for first 2 days (measurements above DL)
- Apart from events: VCs decrease towards the DL whilst leaving the polluted region around Japan
- GOME-2, SCIAMACHY (r<150 km around ship position) larger because of contributing pixels over Japan

Tropospheric profiles:

- VMR < 0.8 ppt even in polluted environment around Japan
- Oct 10 events: Transport from land
- Oct 11 event: Passing other ship
- Apart from events: VMR does not exceed 0.1-0.2 ppb
  → consistent with background estimate of 50 ppt

Fig. 10: GOME-2 trop. NO_2 (October 2009 average)

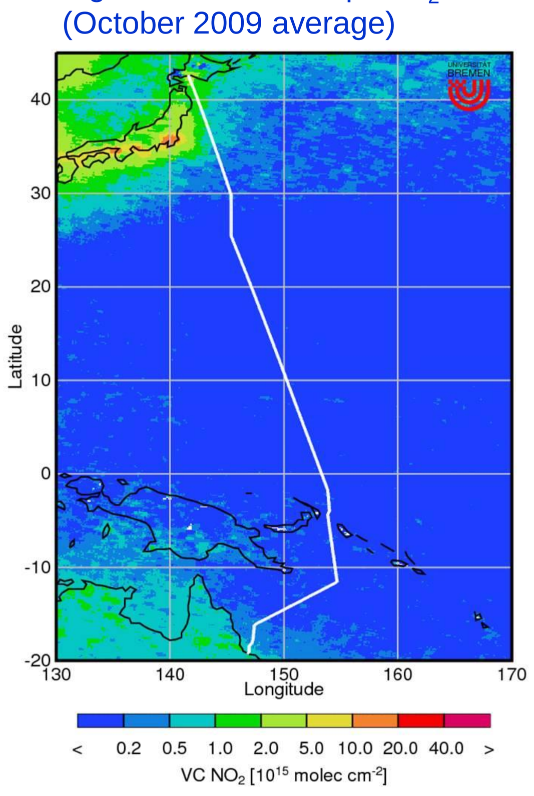
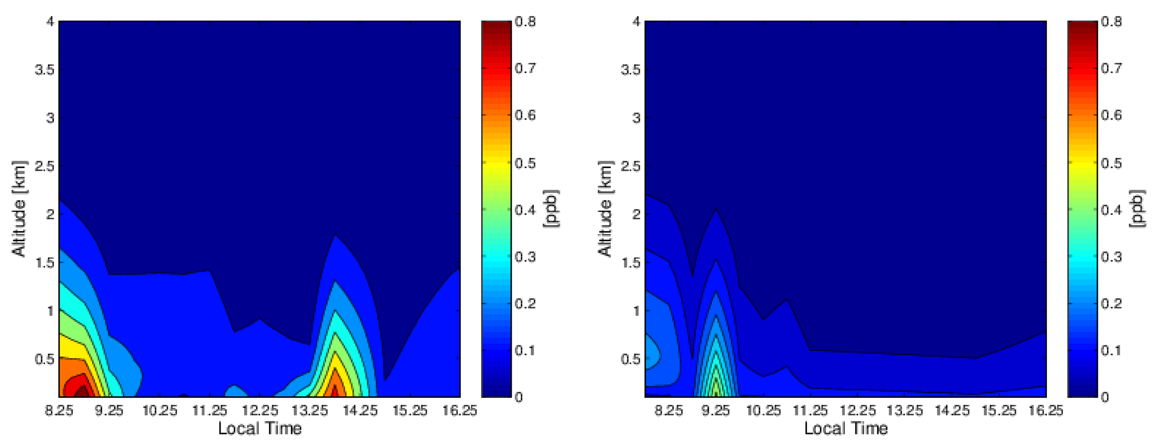
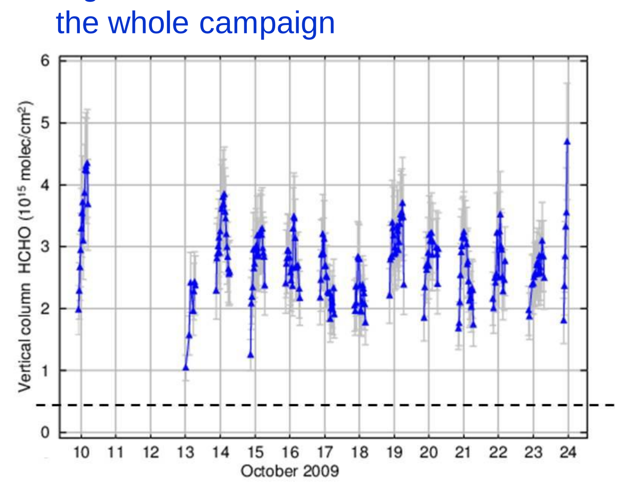


Fig. 9: Tropospheric NO_2 profiles for Oct 10 (left) and 11 (right), close to Japan



Validation III – Formaldehyde

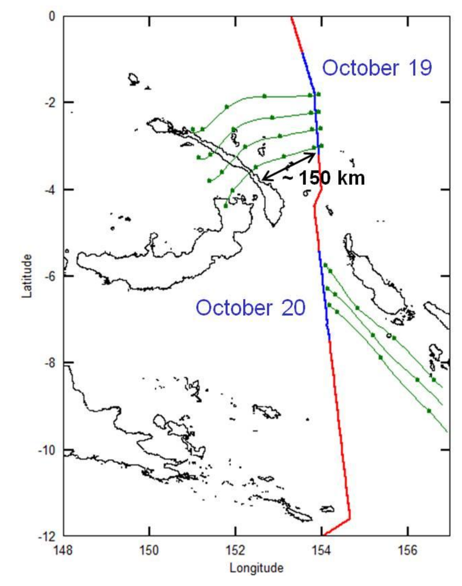
Fig. 11: HCHO vertical columns for the whole campaign



Vertical columns:

- Above DL for the whole cruise
- Far away from sources (e.g. rainforest)
  → background level (mainly from methane oxidation)
- Diurnal cycle
- Oct 10, 24: Influence from land (leaving Japan and arriving in Australia)
- Oct 19: Possibly transport event of HCHO precursors from rainforest island
- Oct 14: Sunniest day of the cruise

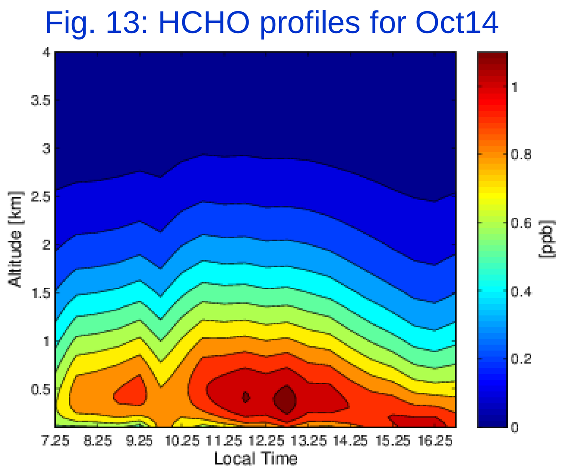
Fig. 12: Cruise Track of Oct 19-20. Green: HYSPLIT backward trajectories



Profiles:

- October 14 case study (maximum in VC timeseries)
- Sunniest day → largest HCHO production (possibly also enhanced HCHO precursors in air masses at 14 Oct, e.g. enhanced DMS measured)
- Clear diurnal cycle
- Maximum (≈1.1 ppb) around noon in elevated altitudes (≈ 400 m)

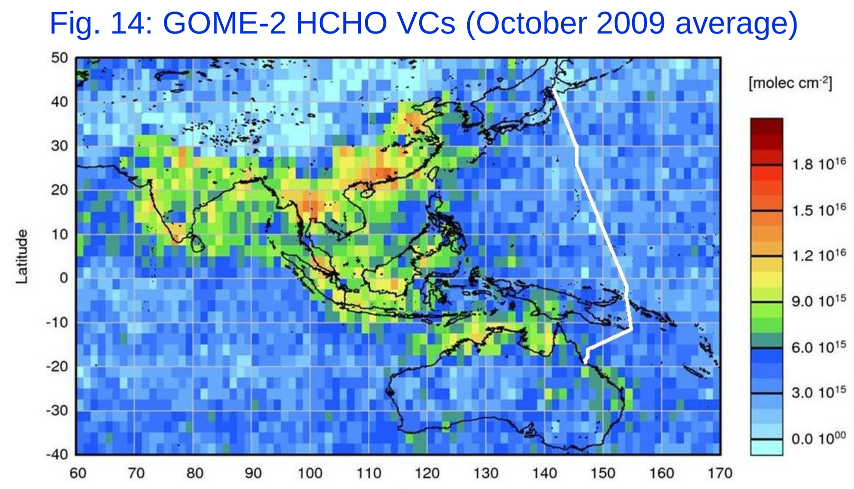
Fig. 13: HCHO profiles for Oct14



Validation (GOME-2 only):

- Uncertainty/scatter of GOME-2 data requires using monthly average
→MAX-DOAS VCs between 9-11 LT (overpass time) averaged and compared to GOME-2 monthly average as function of latitude
- agreement on typical value of ≈ 3 x 10^{15} molec/cm² at overpass time

Fig. 14: GOME-2 HCHO VCs (October 2009 average)



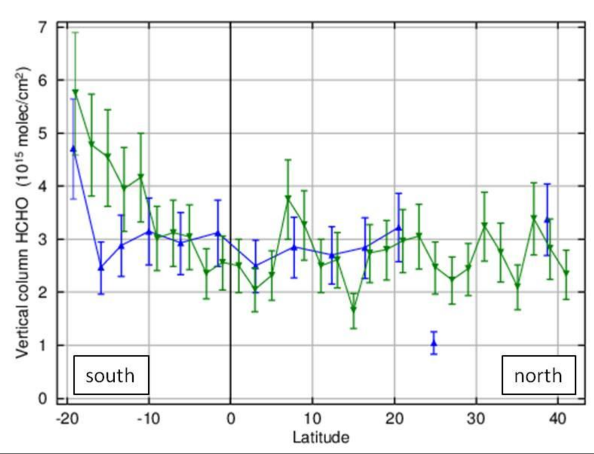


Fig. 15: HCHO VCs as a function of latitude. Green = GOME-2, blue = MAX-DOAS averaged between 9-11 LT (overpass)

Conclusions

- Ship-borne MAX-DOAS measurements of NO_2 and HCHO were performed over the remote western Pacific ocean and vertical columns as well as tropospheric profiles were retrieved.
- The results were used to validate corresponding GOME-2 and SCIAMACHY satellite observations.
- Stratospheric NO_2: GOME-2 and SCIAMACHY differ by 1-1.7% from each other and are on average slightly lower than MAX-DOAS a.m. values, which is a result of the overpass time. From MAX-DOAS, a linear increase of ≈ 8.7 x 10^{13} molec/cm²/h was estimated for stratospheric NO_2 in the tropics.
- Tropospheric NO_2: For the remote ocean a background concentration of ≈ 50 ppt (1.3x10^{14} molec/cm²) was estimated, which is in agreements with the GOME-2 monthly average. Coinciding satellite values were 2-3 times higher, which is an effect of large horizontal averaging (contribution of polluted pixels).
- Formaldehyde: VCs were retrieved above the DL for the whole cruise exhibiting a clear diurnal cycle. Profiles for October 14 having best (sunniest) weather conditions reach maximum concentrations of 1.1 ppb around noon in elevated altitudes (≈400m). MAX-DOAS average values between 9-11 LT and the GOME-2 October 2009 average agree on a typical VC of ≈ 3 x 10^{15} molec/cm² at the overpass time.

Universität Bremen
www.iup.uni-bremen.de/doas

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