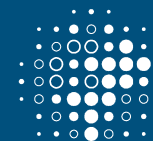


# Observation of clouds with an airborne DOAS instrument

Klaus-Peter Heue<sup>1</sup>, David Walter<sup>1,2</sup>, Carl Brennknecht<sup>1</sup>, Thomas Wagner<sup>1,2</sup>, Tim Deutschmann<sup>2</sup>, Ulrich Platt<sup>2</sup>, Andreas Zahn<sup>3</sup>

- 1) Max-Planck-Institut für Chemie, Mainz, Germany
- 2) Institut für Umweltphysik, Universität Heidelberg, Heidelberg, Germany
- 3) Institut für Meteorologie und Klimaforschung (IMK), Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany

Klaus-Peter.Heue@MPIC.de



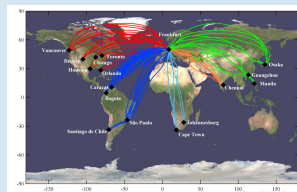
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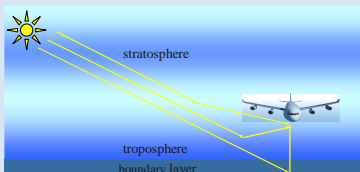
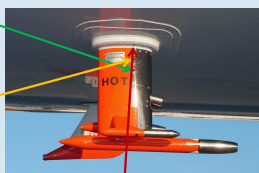
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## Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container

CARIBIC uses a Lufthansa Airbus A340-600 equipped with a dedicated air inlet system. The monthly deployment of the 1.5 ton measurement container on typically 4 intercontinental flights provides regularly detailed data for the UT/LS and tropical free-troposphere. Since December 2004 ~300 flights were successfully performed.



The inlet system has probes for trace gases, for aerosols, and for gaseous and total water. A DOAS system with 3 telescopes (-82°, -10° and +10°) is integrated. For cloud observation a video camera is installed in the inlet system as well.

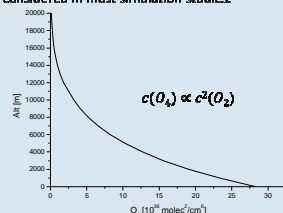


## Observation of clouds with the DOAS instrument (theory)

The DOAS instrument observes scattered or reflected sunlight. The light is spectrally analysed in a wavelength range between 300 and 400nm. Based on Lambert-Beer's law the integrated concentration of several atmospheric absorbers can be retrieved simultaneously.

$$I(\lambda) = I_0(\lambda) \cdot \exp\left(-\sum_{\text{absorbers}} SCD \cdot \sigma(\lambda)\right)$$
$$SCD = \int_{\text{light path}} c(\vec{r}) \cdot d\vec{r}$$

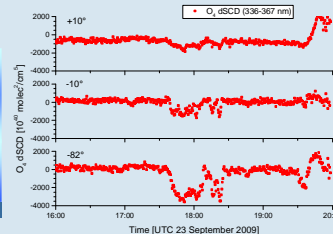
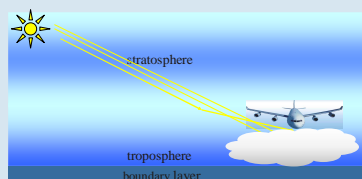
The slant column density (SCD) is the concentration integrated along the light path. Usually the light path through the atmosphere is unknown, therefore tracers with a known concentration are used to estimate the path length. The oxygen dimer  $O_4$  is normally used as it has several absorption features in the UV-vis wavelength range and the concentration is proportional to the square of the  $O_2$  concentration. Also the Ring effect (filling in of the Fraunhofer lines caused by inelastic Raman scattering) contains some information about the number of the scattering events. However, the simulation of the Ring effect requires a radiative transfer simulation in a wide wavelength range, therefore only the  $O_4$  column density and the intensity are considered in most simulation studies.



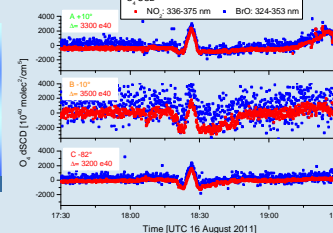
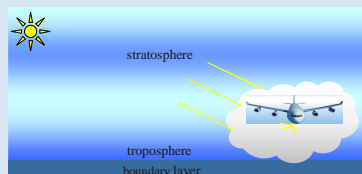
The  $O_4$  concentration is proportional to the square of the  $O_2$  concentration, therefore it decreases exponentially with altitude with a scale height of ~4 km. Hence the variability in time and space is very low. Hence the changes in the  $O_4$  column density are caused by changes in the light path.

## Observation of clouds with the DOAS instrument (example)

The signal of the  $O_4$  dSCD or the Ring effect depends on the altitude of the clouds relative to the aeroplane. If the cloud is below the plane, but rather high, the cloud shields the lower altitudes, this causes a reduction in both the  $O_4$  dSCD and the Ring effect. If the plane is insight a big the light path is extended due to multiple scattering.

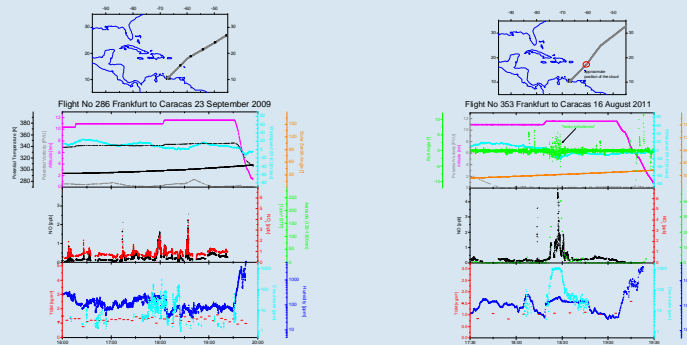


Two examples for the change of the  $O_4$  signal caused by clouds: Above (2009-09-23) on the route between Frankfurt and Caracas the Airbus flew just over or in the top of a cloud, here the  $O_4$  SCD in nadir and -10° are strongly reduced. Also in the +10° viewing direction a decrease in the  $O_4$  signal can be made out. Below (2011-08-16) on the same route the CARIBIC aeroplane penetrated through a large cloud system. In the centre of the cloud a strong increase in the  $O_4$  SCD is observed. The decrease around the peak is caused by the fact, that the plane first passed over parts of the cloud.



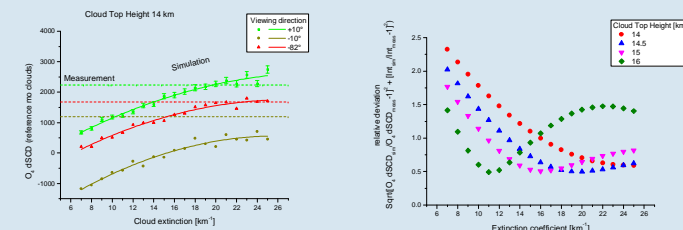
## Other CARIBIC instruments

The video camera in the pylon only showed a white screen for certain period when the plane crossed the cloud. Usually the hull of the plane and the aerosol inlet are visible. In this case the inlet was hidden by the cloud. However the quality of the camera pictures is not sufficient to estimate the cloud optical thickness based on the (in)visibility of the aerosol inlet. The cloud water showed a strong increase during the period, liquid water content of 1100 ppm (0.23 g/m³) was measured. The liquid water content and the assumption of an effective droplet radius of 15-25 µm peak (20 µm) corresponds to an optical extinction of 14 to 25 km⁻¹ and peak value around 18 km⁻¹. For the other example only a cloud content of 200 ppm was measured.



## Radiative Transfer Simulation

To estimate the cloud optical properties, a radiative transfer simulation was performed. The Box Air Mass Factors in the cloud were calculated based on the Monte Carlo based 3D radiative transfer model McArtim (www.iup.uni-heidelberg.de). We assumed a fixed cloud base (1km) and varied cloud top height and cloud optical thickness, the results were compared to the measured  $O_4$  dSCDs and intensity ratios for all 3 viewing directions.

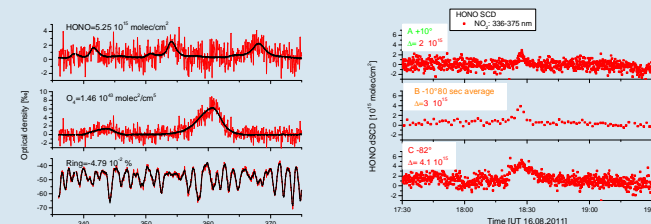


The simulated data were interpolated (left) and the deviation of the simulation to the measurement was calculated (right). For most cloud top altitudes a minimum in the deviation was found. A good agreement between measurements and simulation is found for a cloud top height of 14.5 to 15 km and a cloud extinction of 20 to 15 km⁻¹.

No perfect agreement between the simulated and measured  $O_4$  dSCDs and intensities was found. However, for a cloud top height of 14.5 to 15 km the deviation showed a minimum for the cloud extinction coefficients of 20 or 15 km⁻¹, respectively. This agrees well with the insitu cloud water observation. According to our simulation the total cloud optical thickness was between 210 and 270. The cloud top height (~15km) is also in good agreement with modelled data. TRAJKS an ECMWF based model from KNMI showed a cloud top height of 120 hPa (~15km).

## Observations of HONO in a cloud

Due to the enhanced light path (130-140 km) inside the cloud the sensitivity for certain tracers is enhanced. Therefore HONO, HCHO and NO₂ are observed in the cloud.



Details of the DOAS fit (left, -82° 18:27:05 UTC), the absorption structures of HONO, and  $O_4$  are clearly visible, although the Ring effect is the dominating absorber. Also in the other two viewing directions HONO is observed (right), for -10° the noise is quite high, therefore here only the average of 10 spectra is shown.

With an average light path of around 130 km inside the cloud the measured HONO SCD corresponds to a HONO concentration of 30 ppt. Formaldehyde (0.3 ppb) and NO₂ (0.1 ppb) were measured in the vicinity (7km-CTH) of the aeroplane. Similar observation have been made before e.g. August 1 2008 between Guangzhou and Frankfurt, were a HONO mixing ratio close to 70 ppt was retrieved.

## References:

B. Dix: Spectroscopic Measurements of Atmospheric Trace Gases on Long-Distance Flights, PhD Thesis, University Heidelberg, 2007.  
Heue, K.-P., Brennknecht, C., A. M., Baker, A. K., Baute-Schuch, A., Walter, D., Wagner, T., Hörmann, G., Siller, H., Dü, B., Frick, U., Platt, U., Martinsson, B. G., van Velthoven, P. F. J., Hermann, M., Zahn, A., and Eisinghaus, R.: SO₂ and BrO observation in the plume of the Eyjafjallajökull volcano 2010: CARIBIC and GOMC-2 retrievals, Atmos. Chem. Phys., 11, 2973–2989, doi:10.5194/acp-11-2973-2011, 2011.

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