

# Nitrogene Oxides in the UTLS: Observations from CARIBIC

G. Stratmann<sup>1</sup>, H. Ziereis<sup>1</sup>, H. Schlager<sup>1</sup>, P. Stock<sup>1</sup>, M. Scheibe<sup>1</sup>,  
U. Schumann<sup>1</sup>, F. Slemr<sup>2</sup>, C.A.M. Brenninkmeijer<sup>2</sup>, P. van Velthoven<sup>3</sup>, A. Zahn<sup>4</sup>

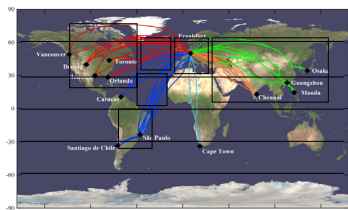
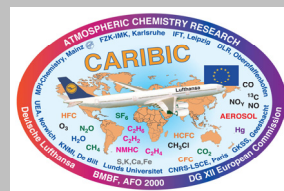


Fig. 1: Flight routes of CARIBIC ([www.caribic-atmospheric.com](http://www.caribic-atmospheric.com))

## 1. NO and NO<sub>y</sub> Measurements on CARIBIC

A unique set of airborne in situ observations of nitrogen oxide (NO) and the sum of all reactive nitrogen species (NO<sub>y</sub>) has been performed in the tropopause region. These data have been acquired within the CARIBIC project (Civil Aircraft for the Regular Investigation of the Atmosphere Based on an Instrument Container, ([www.caribic-atmospheric.com](http://www.caribic-atmospheric.com)). Since December 2004 NO and NO<sub>y</sub> data have been obtained on a monthly base during more than 170 flights using a Lufthansa Airbus A340-600 (Fig.1). More than 2 250 000 NO<sub>y</sub> data points were measured during the CARIBIC program to date.

Nitrogen oxides play a key role in atmospheric photochemistry, particularly in controlling the cycling of OH and the production of ozone in the upper troposphere and lower stratosphere (UTLS). The budget of nitrogen oxides in the UTLS is controlled by a variety of different sources and processes, chiefly: long-range transport, lofting from the boundary layer, lightning, anthropogenic pollution from industry and air traffic emissions.

In this study the nitrogen oxide data are analyzed along regional and seasonal differences, along with species as CO. The large scale distribution of NO and NO<sub>y</sub> in the UTLS is presented. Tracer correlations are used to investigate the contribution of biomass burning on the nitrogen budget. Attempts have been made to assess variations in tropospheric and stratospheric influenced samples of nitrogen oxides.

## 2. Seasonal NO<sub>y</sub> Variability in Different Regions

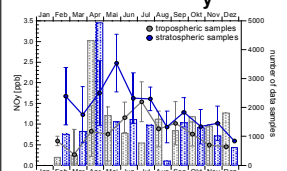


Fig. 2.1: Seasonal Cycle over Europe (12°W-30°E, 35°-63°N)

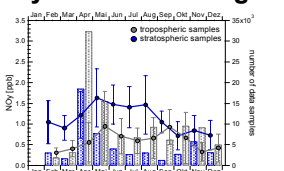


Fig. 2.2: Seasonal Cycle over Asia (30°-140°E, 10°-65°N)

The seasonal variation of NO<sub>y</sub> concentration in different regions is shown in Fig.2.1-2.3. The classification in tropospheric and stratospheric samples results from the potential vorticity (PV<2 → tropospheric; PV>2 → stratospheric). The bars denote the number of data samples. On average measurements were performed at an altitude of about 33 100 ft.

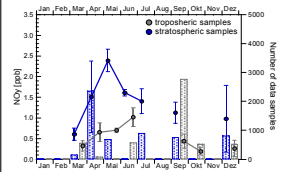


Fig. 2.3: Seasonal Cycle over the North Atlantic (12°-53°W, 30°-65°N)

## 3. Longitudinal NO<sub>y</sub> Distribution at midlatitudes (30°-60°N) - Seasonal Variaton

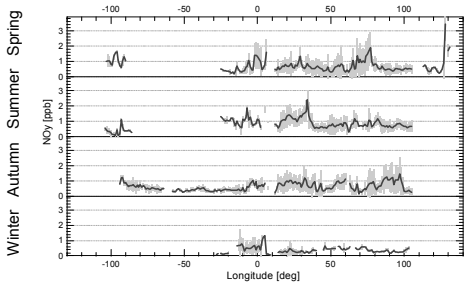


Fig.3.1: tropospheric samples

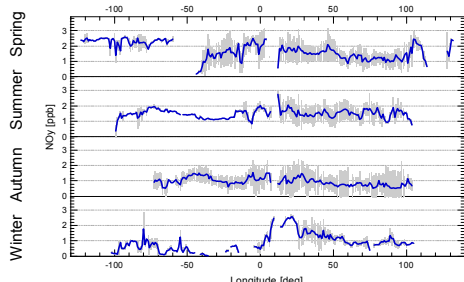


Fig.3.2: stratospheric samples

Between May 2004 and July 2009 167 missions were performed at northern midlatitudes. Nearly 58% of all CARIBIC NO<sub>y</sub> data were obtained in this area. Fig. 3.1 and 3.2 contain about 1 300 000 NO<sub>y</sub> data samples. These missions included flight routes to Asia (Guangzhou, Chennai, Osaka and Manila), North America (Denver, Houston, Vancouver, Toronto, Orlando) and South America (Caracas, Buenos Aires, Sao Paulo and Santiago de Chile). NO<sub>y</sub> is averaged over longitude bins of 1°. The standard deviation is shown in light grey.

## 4. Case Study - Indication of Biomass Burning

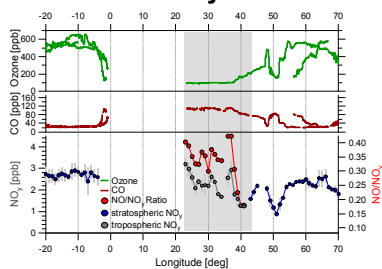


Fig. 4.1: longitudinal distribution of NO<sub>y</sub> in the polar region (60°-80°N) in spring

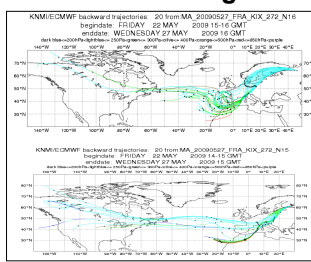


Fig. 4.3: five day backward trajectories (Peter van Velthoven, KNMI Netherlands)



Fig. 4.4: MODIS Firemap from May 21 till may 30 (<http://rapidfire.sci.gsfc.nasa.gov/firemaps/>)

Tracer correlations of NO<sub>y</sub> with species as CO, O<sub>3</sub>, aerosol particles and others can be used to investigate the contribution of different sources to the nitrogen budget. In May 2009 high tropospheric NO<sub>y</sub> values were observed between 20°- 40°E and 60°- 80°N (respectively fig. 4.1). Additional a high NO/NO<sub>y</sub> ratio was found. A high ratio can be an indicator for fresh polluted air. By correlating NO<sub>y</sub> with CO a strong signature

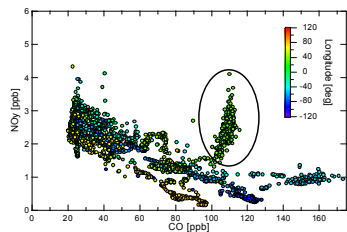
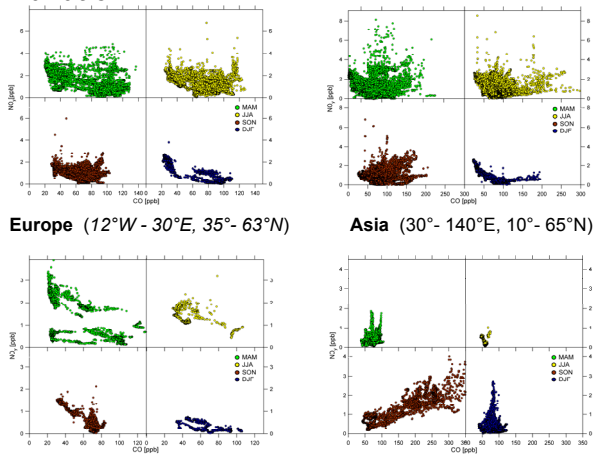


Fig. 4.2: NO<sub>y</sub> versus CO for CARIBIC data samples in the polar region

becomes apparent (fig. 4.2). Back trajectory calculations showed that the air mass sampled originated from North America 3-5 days earlier. MODIS (Moderate Resolution Imaging Spectroradiometer) firemaps from May 2009 show that the air mass might be affected by biomass burning events in North America. For a doubtless identification of the origin of these air masses further analysis including aerosol measurements are necessary. Also the meteorological situation has to be investigated.

## 5. NO<sub>y</sub>-CO Correlations - Regional and Seasonal Variation



Europe (12°W - 30°E, 35°- 63°N) Asia (30°- 140°E, 10°- 65°N)  
North Atlantic (12°- 53°W, 35°-65°N) South America (30°- 70°W, 3°- 35°S)

## 6. Outlook

- Detailed investigation of the large scale NO<sub>y</sub> distribution at the UTLS
- Investigation of sources contribute to the NO<sub>y</sub> ratio at the UTLS
- Study of the background NO<sub>y</sub> enhancement caused by air traffic
- Comparison with model simulations and other measurements, e.g. MOZAIC
- Installation of a NO<sub>2</sub> Converter additional to the NO and NO<sub>x</sub> Converter

<sup>1</sup>Deutsches Zentrum für Luft- und Raumfahrt, Physik der Atmosphäre, Oberpfaffenhofen, Germany; <sup>2</sup>Max Planck Institute for Chemistry, Atmospheric Chemistry Division, Mainz, Germany; <sup>3</sup> KNMI Royal Dutch Meteorological Institute, de Bilt, the Netherlands; <sup>4</sup> Institute for Meteorology and Climate Research, Forschungszentrum Karlsruhe, Karlsruhe, Germany