Annual Report 2001

for the AFO2000-Project

"Convective Transport of Trace Gases into the Upper Troposphere over Europe: Budget and Impact on Chemistry" (CONTRACE)

FKZ: 07 ATF 19 - 22

by

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Project Period: 01.01.2001 – 31.12.2003

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"Convective Transport of Trace Gases into the Upper Troposphere over Europe: Budget and Impact on Chemistry" (CONTRACE)

Annual report 2001 by „Gesamtverbund" (DLR, MPI-K, IFU and TUM)

- **Kick-off-Meeting, 15-16th March 2001 in Oberpfaffenhofen.**

The upcoming field experiment in November 2001 was discussed and planned in detail (development of instruments, integration in the Falcon, forecast products for the briefing). Further, a collaboration with two external CONTRACE partner, H. Elbern (EURAD-Köln) and M. Lawrence (MPI-Mainz), was arranged. H. Elbern will contribute with meteorological and chemical forecast products for the planning of the flights and will use the CONTRACE flight measurements for 4D data assimilations (another AFO2000-project). M. Lawrence will provide chemical transport model calculations for the field experiment phase.

- **A web page was created for CONTRACE.**

http://www.pa.op.dlr.de/contrace

- **Technical meeting, 12th October 2001 in Oberpfaffenhofen.**

Planning of the instrument integration on the Falcon.

- **First airborne field experiment, 5-30th November 2001 in Oberpfaffenhofen.**

The Falcon was equipped with an extensive set of instruments to measure CO, CO_2, O_3, NO, NOy, $J(NO_2)$, water vapour, $J(O^1D)$, nonmethane hydrocarbons, formaldehyde, hydrogen peroxide, FSSP, SO_2, acetone, methanol, HNO_3 and ions by DLR, IFU and MPI-K. A large set of meteorological and chemical forecast products were provided by TUM, EURAD and MPI-Mainz for the daily briefing (see http://www.pa.op.dlr.de/contrace/). For the first time chemical forecast products were used to guide the Falcon directly into polluted air masses in the mid and upper troposphere which turned out to be very useful. The chemical forecast models run by TUM and MPI-Mainz can further separate the vertical transport of European and North American surface emissions (CO used as tracer). About once a week it was observed that polluted North American air masses moved over Europe. The backward trajectories showed how frontal systems with pronounced *warm conveyor belts* moved over the polluted eastern USA and lifted air from the boundary layer. If these emissions attained the jet stream they were transported very rapidly (within days) to high altitudes over Europe. During the four mission flights (to Corsica, Stockholm, Budapest, and Brussels) polluted layers with both European and North American surface emissions were successfully found in the middle and upper troposphere over Europe. Enhanced CO, CO_2, NO, NOy, and SO_2 mixing ratios were observed in these layers.

- **CONTRACE publications to intercontinental transport and convective mass flux:**

Stohl, A., S. Eckhardt, C. Forster, P. James, and N. Spichtinger, On the pathways and timescales of intercontinental air pollution transport, submitted to *J. Geophys. Res.*, October 2001.

Huntrieser et al., CONTRACE - Convective Transport of Trace Gases into the Middle and Upper Troposphere over Europe, submitted to *EUROTRAC Symposium* 2002.

Wimmer, S., Entwicklung eines Algorithmus zur Bestimmung des vertikalen Massenflusses in einzelnen hochreichenden Konvektionszellen aus Meteosat-Daten, Diplomarbeit abgeschlossen Februar 2002, Universität München.

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"Convective Transport of Trace Gases into the Upper Troposphere over Europe: Budget and Impact on Chemistry" (CONTRACE)

Annual report 2001 by Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre

H. Huntrieser, H. Schlager, J. Heland, H. Mannstein and S. Wimmer

The first airborne CONTRACE field experiment was carried out in November 2001. The tasks of the DLR were to coordinate the campaign, to plan the flights and to perform measurements (CO, CO_2, O_3, NO, NOy, J(NO_2) and water vapour) on the Falcon. The first field phase was successful for DLR since all instruments run without any problems. On the basis of the chemical forecast products (provided by TUM, EURAD and MPI-Mainz) it was possible for the first time to direct the Falcon into polluted air masses (European or North American origin). It was observed that in the winter time mainly large scale lifting along cold fronts (so-called *warm conveyor belts*) was responsible for the transport of surface emissions to the free troposphere. During all four mission flights (to Corsica, Stockholm, Budapest, and Brussels) polluted layers were successfully found in the mid and upper troposphere. Enhancements due to the emissions were observed mainly in the CO, CO_2, NO, and NOy mixing ratios. In Figure 1 an example from 19th November is presented. The chemical model forecasts by TUM and MPI-Mainz showed an extended layer of CO emissions with North American origin over Scandinavia. The measured CO profiles over Oslo and Stockholm also distinctly showed the polluted layer with CO mixing ratios up to 160 ppbv.

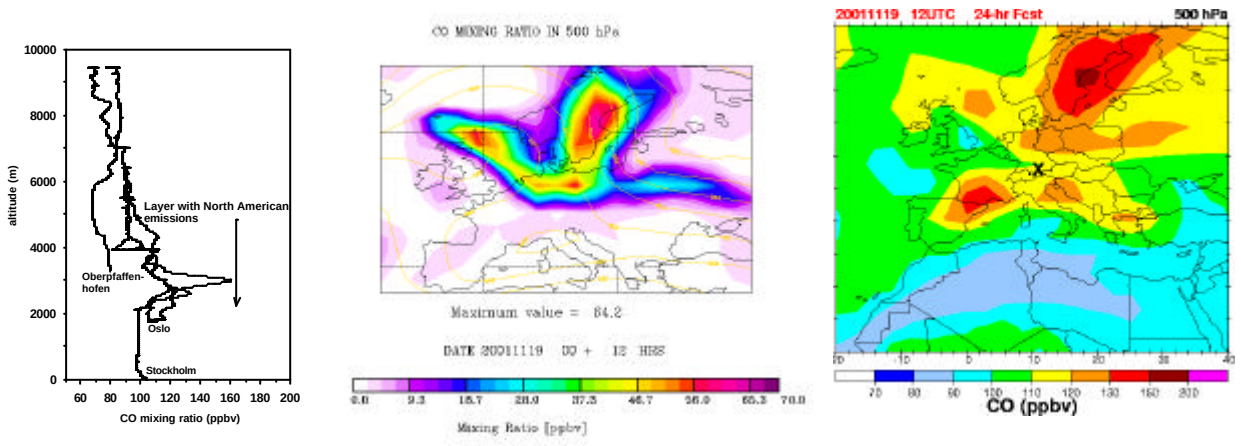

Figure 1: *Vertical CO-profiles from 19th November and predicted CO distributions (TUM and MPI-Mainz).*

Parallel to the planning and performance of the first CONTRACE field experiment a new algorithm for detection and tracking of convective clouds in the Meteosat images was developed. First tests of the algorithm and statistical analyses (growing rate of the clouds, frequency of the clouds over Europe, preferred pathways) were carried out and compared to other sources (for example lightning statistics).

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"Convective Transport of Trace Gases into the Upper Troposphere over Europe: Budget and Impact on Chemistry" (CONTRACE)

Sub-project: Upper tropospheric peroxy radicals

Annual report 2001 by MPI für Kernphysik, Bereich Atmosphärenphysik

F. Arnold, H. Aufmhoff, S. Eichkorn and S. Wilhelm

The first aircraft measurements took place in November 2001 at DLR, Oberpfaffenhofen. On 4 flight days, the trace gases SO_2, acetone (an important peroxy radical precursor), methanol, and HNO_3 were measured with the CIMS instrument (Chemical Ionization Mass Spectrometer), as well as the mass distributions of positive and negative massive natural atmospheric (formed by galactic cosmic radiation) ions with the LIOMAS instrument (Large-Ion Mass Spectrometer), and the total ion concentration with 2 electrostatic probes (one outside the aircraft, the other within the flow tube). A new permeation source enabled in-flight calibrations not only for SO_2, but also for acetone and methanol. Apart from minor technical problems during the first test flight, all instruments worked very well.
The preliminary results show that convective pollution events were found in the upper troposphere. Figure 2 shows that the SO_2 signal has increased threefold from time to time on the last flight level (~5800 m). The HNO_3 signal does not follow these strong changes, but the CO signal measured by DLR correlates with them. To arrive at a final data correction, we are presently investigating the dependence of the measured signals on the atmospheric water vapour concentration.

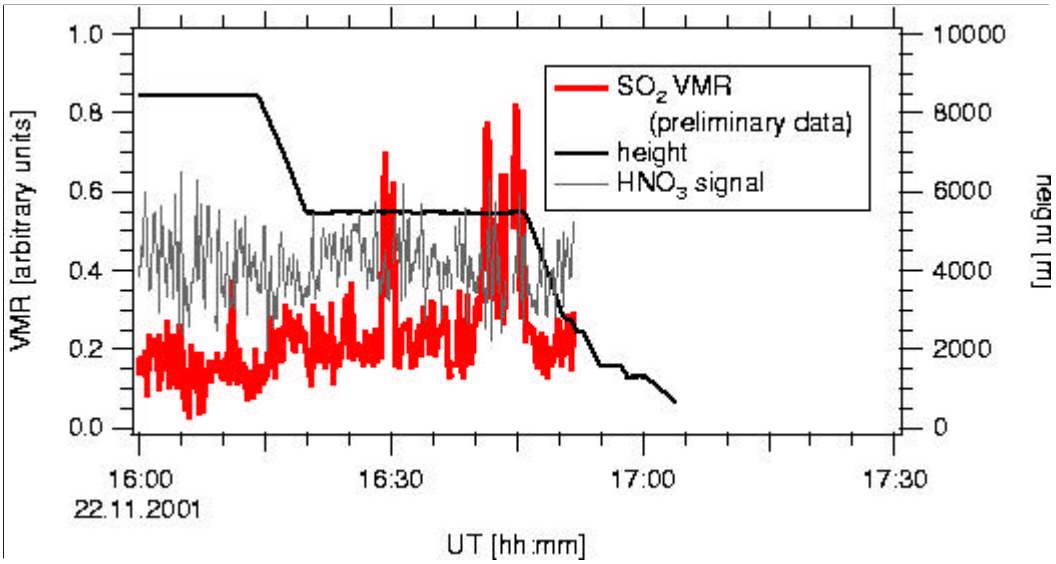

Figure 2: *Concentrations of SO_2 and HNO_3 (preliminary data) together with altitude vs. time on 22.11.01*

Previous investigations in summer (S. Eichkorn, PhD Thesis, Heidelberg 2001) showed that occasionally, particularly in polluted, convectively uplifted air masses, strong ion growth takes place in the upper troposphere. With these events, the maximum mass of positive ions increased from about 400 amu to 600-700 amu and sometimes even up to 2500 amu. These massive ions are precursors of aerosol particles. Such processes were to be investigated for comparison also in winter, measuring for the first time positive and negative ions simultaneously. The preliminary data points to a weaker variability of the mass distribution of negative ions compared to positive ions.

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"Convective Transport of Trace Gases into the Upper Troposphere over Europe: Budget and Impact on Chemistry" (CONTRACE)

Annual report 2001 by IFU, Garmisch-Partenkirchen

W. Junkermann, H. Giehl and R. Meier

Within the project measurements of the photolysis rate of ozone (JO1D), nonmethane hydrocarbons in grab samples, formaldehyde and hydrogen peroxide were foreseen. While the instrumentation for radiation and grab sampling was available, new instruments had to be built for the trace gases. The technique for both instruments is fluorimetry in the liquid phase after stripping the gas phase compounds in a stripping coil. In the laboratory for both compounds detection limits below 50 ppt were achieved at a time resolution between 60 and 90 sec. As the technique is sensitive to pressure differences the whole instrument has to be kept either on ambient pressure or on cabin pressure in a pressurized aircraft. As keeping the instruments at ambient pressure proved to be very sensitive to leaks in previous flight experiments a pressurized inlet system was designed from inert material using a membrane compressor. This approach allows to operate the instrument within the cabin at cabin pressure but needs a careful design and characterization to prevent from losses or artifact produced by the inlet system. The characterization has to cover all possible temperature, humidity and pressure ranges during the proposed flight missions. We found no detectable differences for formaldehyde and a maximum of five percent loss for hydrogen peroxide compared to an unpressurized inlet system of the same length / residence time. The formaldehyde instrument was field tested in a precampaign on the French MERLIN aircraft and proved to need an updated fluorimeter as the old version was not stable enough.

The instruments were used during the November campaign and for this campaign a Forward Scattering Spectrometer Probe (FSSP) was added to the payload as this instrument allows to characterize droplet spectra for conditions within clouds were chemical losses are expected for the water soluble compounds.

The instruments were operated successfully as long as the flights started from the Oberpfaffenhofen base. The pressurized inlet system worked fine for altitudes up to 31000 ft but was unable to deliver enough air above. Unfortunately we had severe problems with the formaldehyde system after each new take off when the flight was interrupted. The instrument was contaminated and needed about two hours of cleaning to get back into normal operating conditions. Different measures were taken to prevent contamination including backflushing of the inlet with clean air etc. but the problem could not be solved completely. Thus further investigations have to be done in 2002. For the next campaign a more powerful compressor will be used as flight altitudes will reach up to 37000 ft. All other instruments were running properly.

The results proved the expected low values of the formaldehyde and peroxide mixing ratios in the fall / winter high troposphere.

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Annual report 2001 by Technische Universität München (TUM),
Lehrstuhl für Bioklimatologie und Immissionsforschung
C. Forster and A. Stohl

1. Objectives for the reporting period

- Support of the flight planning during the airborne field campaigns by providing prognostic model forecasts
- Establish a warm conveyor belt (WCB) climatology

2. Most important results for the reporting period

During the airborne field campaign in November 2001 TUM provided different forecast products on a webpage (see http://www.fw.tum.de/EXT/LST/METEO/contrace_fx.html). The forecasts cover a period of 3 days and were updated every 6 hours during the campaigns, every 24 hours otherwise. They are based on AVN data, since the German Weather Service did not yet give a permission for the use of the ECMWF prognostic fields. Meteorological fields (geopotential, tropopause height, surface pressure, temperature, relative humidity, equivalentpotential temperature, precipitation and cloud cover) are provided as well as trajectory and tracer forecasts, which are calculated with the models FLEXTRA and FLEXPART, respectively. FLEXPART calculates the transport and distribution of a North American anthropogenic and a European anthropopogenic CO tracer based on the EDGAR emission inventory (Olivier et al., 1996). This makes it possible to identify the transport patterns and the source region of a polluted air mass. Figure 3 shows an example for a tracer and trajectory forecast.

A WCB climatology over 15 years has been established with FLEXTRA. The seasonal and geographic variations of WCB frequency and intensity were investigated.

3. Deviations from the work plan

There were no deviations from the work plan.

4. References

Olivier, J. G. J., et al., Description of EDGAR Version 2.0. A set of globel emission inventories of greenhouse gases and ozone depleting substances for all anthropogenic and most natural sources per country basis and on 1x1 grid. RIVM/TNO report, December 1996.

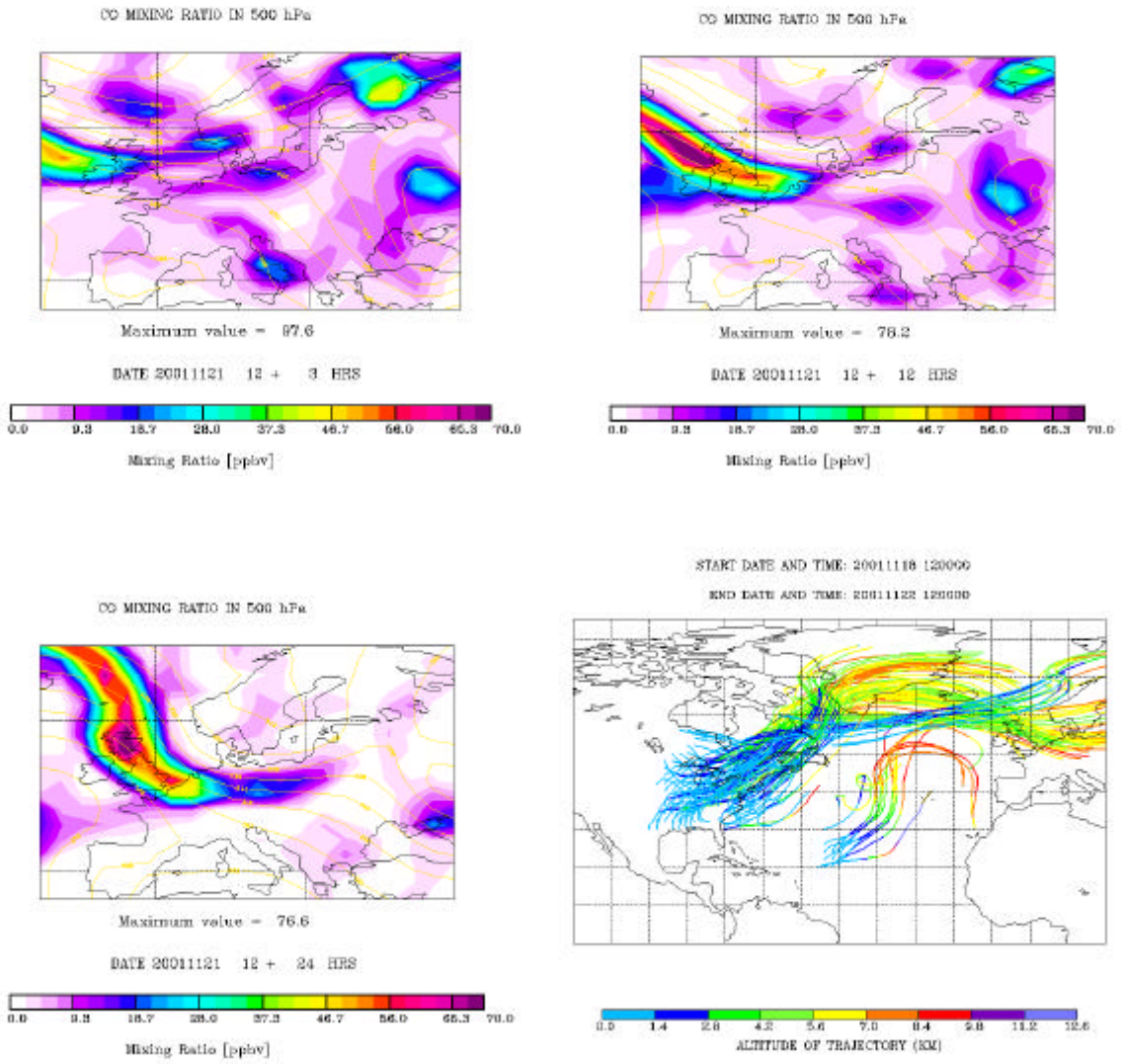

Figure 3: *Upper panels and lower left panel: FLEXPART forecast of the mixing ratio of a North American CO tracer at 500 hPa in combination with the geopotential height (in gpdm, yellow contours). A North American plume moves from the Atlantic over Britain to the northern part of Germany. Lower right panel: 4-days FLEXTRA forward trajectories, which fulfill a warm conveyor belt criterion (ascent over more than 5 km altitude in 4 days, endpoint of trajectory NE of startpoint), started in the domain -110°-20° E and 20°-60°N. The trajectories show transport of North American boundary layer air to Europe within a warm conveyor belt. The ending time of the trajectories corresponds to the time in the lower left panel.*