<u>Annual Report 2002</u>

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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Annual report 2002 by „Gesamtverbund" (DLR, MPI-K, FZK and TUM)

- **CONTRACE-Workshop, 2-3 April 2002 in Oberpfaffenhofen.**

The first CONTRACE field experiment was successfully performed. An extensive set of chemical weather forecasts were used to guide the Falcon, for the first time, into polluted air masses that originated over North America (NA) (Lawrence et al., 2002; Stohl et al, 2002). It was observed that these NA-plumes move much more frequently over Europe than expected (weekly) and as a result even distinctly impact the ozone distribution (25% increase in a layer). The extension of these plumes was impressive (1000-2000 km long, 200-300 km wide and 1-2 km deep). FLEXPART backward modeling studies were used to identify the source region of the polluted NA plumes (Stohl et al., 2002). The airborne CONTRACE measurements are the first one which indicate that NA-plumes even strongly influence the ozone distribution over Europe in winter.

- **Response to the "Gutachtervotum" (AFO2000-Statusseminar Schliersee).**

As a response to the "Gutachtervotum" it was decided to slightly change the instrument pay load on the Falcon for the second CONTRACE field experiment. For the new objectives a rack with an extensive aerosol instrumentation will be integrated (DLR). Because of the lack of space no SO_2 and acetone measurements can be performed anymore (MPI-K).

- **Cooperation with other groups (within and outside of AFO2000).**

In the past two years several external partners have joined the CONTRACE-"Verbund": H. Elbern & H. Jakobs (EURAD, Köln), M. Lawrence & H. Fischer (MPI-C, Mainz), T. Trickl (IMK, Garmisch), M. Schultz (MPI-M, Hamburg), A. Richter (IUP, Bremen) and O. Cooper (NOAA, Boulder, USA).

H. Elbern & H. Jakobs produce meteorological and chemical forecasts (EURAD-model) for the flight planning and will use the airborne CONTRACE measurements for 4D-data assimilations (**SATEC4D-AFO2000**). M. Lawrence provides chemical transport model calculations (MATCH-model) for the planning of the flights (**SAPHIRE-AFO2000**). Further it is planned to coordinate the second CONTRACE field experiment with the **EU-project UTOPIHAN** (H. Fischer), joined flights with the Falcon and a Lear-Jet will be performed. T. Trickl has agreed to run the ozone lidar in Garmisch during the second CONTRACE field phase (**ATMOFAST-AFO2000**). M. Schultz plans to run ECHAM simulations for CONTRACE-1 (comparison ECHAM with FLEXPART) and will provide ECMWF forecasts for CONTRACE-2. A. Richter has provided GOME-NO2-images for CONTRACE-1 (**NOXTRAM-AFO2000**, Stohl et al., 2003) and will make current GOME/(SCIA)-NO2 images available for the flight planning of CONTRACE-2. O. Cooper provides satellite composits (METEOSAT-GOES).

Furthermore, for the analysis of some CONTRACE flights we are in contact with colleagues from **SPURT-AFO2000** (H. Fischer) and **CARIBIC-AFO2000** (F. Slemr) since some of the airborne measurements were performed in the same polluted air masses, however at different locations.

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

See reference lists of the individual sub-projects and http://www.pa.op.dlr.de/contrace/.

FKZ: 07 ATF 19

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

Annual report 2002 by Deutsches Zentrum für Luft- und Raumfahrt (DLR), Institut für Physik der Atmosphäre

H. Huntrieser, H. Schlager, R. Busen, C. Hauser, J. Heland, H. Mannstein and S. Wimmer

The detailed analysis of the airborne measurements from the first CONTRACE field experiment has begun. Comparisons between the measured CO data and two different CO model forecasts/analyses (MATCH and FLEXPART) were performed and documented in two publications (Lawrence et al., 2002; Stohl et al., 2002). It was shown that the observed episodes with high CO concentrations in the free troposphere over Europe were mainly caused by CO emissions transported all the way from North America (NA).

Furthermore, the trace gas compositions and correlations in the NA-plumes were analysed (Huntrieser et al., 2002a-b). Besides distinct increases in the CO concentration (up to 170 ppbv) also enhanced NOy values were measured (1-2 ppbv). The NO and NO_2 concentration (=NOx) did not show any enhancement in the NA-plumes (life time too short, already converted to NOz=NOy-NOx). This means that the influence of these aged plumes from NA on the NOx distribution in the free troposphere over Europe is negligible and that no more ozone is produced over Europe (important for the objectives of CONTRACE). However, the correlation analyses show a positive CO-O_3 correlation in the NA-plumes (Fig. 1). This means that ozone production has taken place. For the season of the measurements (winter) a surprising result. The ozone concentration in the NA-plumes (45-55 ppbv) was about 10 ppbv (25%) enhanced in comparison to the background. Highly resolved backward trajectory calculations were used to estimate the origin area of the emissions (Stohl et al., 2002). In this area over the eastern US ozone measurements on mountain stations were analysed. Every time polluted air masses were lifted ahead of an approaching cold front enhanced ozone concentrations (up to 60 ppbv) were measured on top of these stations (in November normally 20-30 ppbv is observed). Probably the ozone production took place already in the highly polluted boundary layer over eastern US. Certain weather conditions, a stagnating high pressure system and almost no precipitation for a long period of time, caused an accumulation of anthropogenic emissions. Considerable amounts of ozone could be produced in these air masses.

Furthermore, the analyses of the airborne measurements show a distinct positive CO-NOy correlation in the NA-plumes. From the CO-NOy slope it is possible to estimate the age of the emissions, in this case mainly between 4-5 days. The estimates from the measurements show a close agreement with the modeled age spectra (Stohl et al., 2002).

At the beginning of the CONTRACE field phase an unusual NA-plume was discovered, which was transported to Europe in only 1-2 days. Due to a certain weather situation this trace gas transport could for the first time be tracked in GOME-NO_2 satellite images (Stohl et al., 2003).

In the frame work of CONTRACE a new algorithm for detection and tracking of convective clouds in the METEOSAT images has been developed. First tests of the algorithm and statistical analyses (growth rate of the clouds, frequency of the clouds over Europe, preferred pathways) have been carried out and further documented (Wimmer, 2002).

a) b)

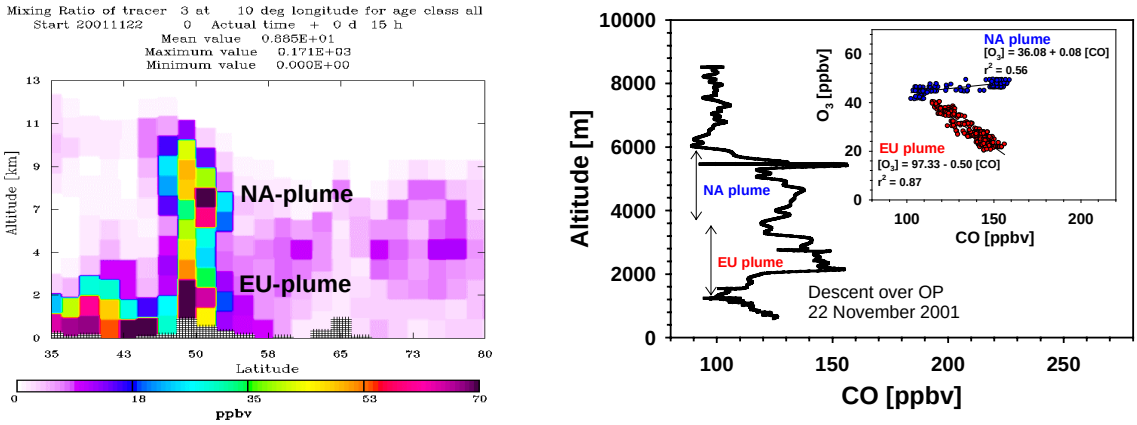

Figure 1: *Vertical cross section of the FLEXPART CO-tracer forecast for 22nd November 2001 (a). Two polluted layers with different origin (European and North American) were located over Germany. During the descent over Oberpfaffenhofen (Germany) the Falcon aircraft penetrated both of these layers (b). In the North American (European) plume a positive (negative)* O_3*-CO correlation was observed.*

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Stohl, A., C. Forster, S. Eckhardt, H. Huntrieser, J. Heland, H. Schlager, H. Aufmhoff, F. Arnold, and O. Cooper, 2002: A backward modeling study of intercontinental pollution transport using aircraft measurements, *J. Geophys. Res.*, in press.

Stohl A., H. Huntrieser, A. Richter, S. Beirle, O. Cooper, S. Eckhardt, C. Forster, P. James, N. Spichtinger, M. Wenig, T. Wagner, J. Burrows, and U. Platt, 2003: Air pollution taking an intercontinental express highway in a bomb, in preparation for *Atmos. Chem. Phys.*

Wimmer, S., Entwicklung eines Algorithmus zur Bestimmung des vertikalen Massenflusses in einzelnen hochreichenden Konvektionszellen aus METEOSAT-Daten, *Diplomarbeit am Institut für Geographie*, LMU München, 28. Februar 2002.

FKZ: 07 ATF 20

"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 2002 by MPI für Kernphysik, Bereich Atmosphärenphysik

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

Mass distribution- and concentration measurements of atmospheric ions

Intensive analysis of the aircraft based ion measurements begun this year. Special focus was set on the investigation of the ion growth performance in polluted air masses. CO was used as pollution tracer. A clear correlation was found between polluted air masses and the abundance of large ions. A simple kinetic model was used to approximate the growth of negative and positive ions by the attachment of condensable gases like gaseous sulfuric acid, which should be more abundant in polluted air masses. CO was used as pollution tracer to calculate empirical growth factors for several mass bins within the whole mass range of the LIOMAS instrument. Mass distributions calculated with this pollution- growth factors show good agreement with spectra during different flights. Figure 1a shows the calculated mass distributions together with two measured spectra taken in air masses with different CO concentrations.

Trace gas measurements of Acetone and Sulfur Dioxide

Together with calibration measurements performed in the laboratory the Analysis of the acetone and sulfur dioxide measurements have been recalibrated within this year. The detection limit was found to be > 0.5ppbV. A good correlation for acetone and CO was found during all flights. A correlation between SO_2 and CO was not detected during all episodes, when air masses with high CO concentrations where encountered. Calculations of acetonitril concentrations from the mass spectrometric data were performed. Thereby a detailed analysis of the pollution sources is possible. In particular acetonitril can be used as a tracer for boreal fires. Figure 1b shows concentrations of acetone and SO_2 during the flight on 22 November 2001. The increase in concentrations of both trace gases where by the trajectory analyses identified as North American pollutions.

(a) (b)

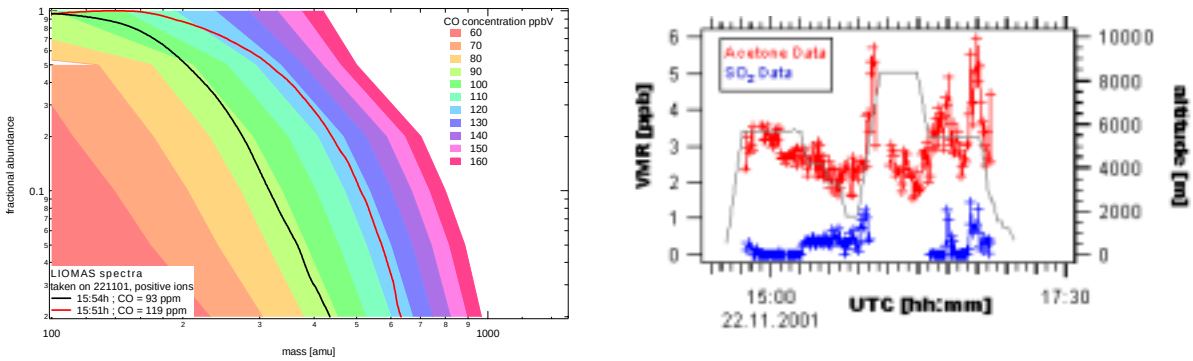

Figure 1: *Ion mass distributions for different CO concentrations. The coloured areas show mass distributions derived with the measured growth factors. The solid lines show measured spectra. (a) . Concentrations of acetone ads SO_2 on 22 November 2001.*

FKZ: 07 ATF 21

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

Annual report 2002 by IMK-FZK, Garmisch-Partenkirchen

W. Junkermann and B. Rappenglück

FKZ: 07 ATF 22

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

Annual report 2002 by Technische Universität München (TUM), Lehrstuhl für Bioklimatologie und Immissionsforschung

C. Forster and A. Stohl

1. Objectives for the reporting period

- Backtrajectory calculations along the flight tracks
- Calculation of tracer age spectra
- Create a warm conveyor belt (WCB) climatology; NO_x transport in WCBs

2. Most important results for the reporting period

Forward simulations of CO and NO_x tracers originating from anthropogenic and biomass burning sources were performed by using the dispersion model FLEXPART (Stohl et al., 2002b). In two case studies WCBs were identified as a mechanism that can transport pollution from North America to Europe on a time scale of about 5 days. The model age spectra and the chemical composition of the trace gas concentrations measured along the flight track confirm this time scale. Mainly emissions from the North American boundary layer and to a lesser extent biomass burning emissions contributed to the measured trace gas concentrations. FLEXPART backward simulations along the flight tracks in combination with an emission inventory allowed a detailed source analysis of the trace gas concentrations measured along the flight tracks (Stohl et al., 2002b). While the east coast of the US and especially New York were the main pollution source in the first case study, the pollution originated in California, Texas and Florida in the second case study (Figure 1). The backward simulations can be regarded as a replacement for conventional backtrajectory calculations (Stohl et al., 2002a).

The 15-year WCB climatology with the trajectory model FLEXTRA has been summarized and submitted to J. Climate (Eckhardt et al., 2002).

Based on NO_2 satellite measurements and FLEXPART simulations a trans-Atlantic pollution transport event on a time scale of about one day has been documented for the first time. The transport took place in a so-called "bomb", which is a rapidly developing exceptionally strong cyclone. A transport climatology revealed that intercontinental transport of trace gases on a time scale of one to two days may affect the budget of short lived trace gases in the remote troposphere.

3. Deviations from the work plan

The second measurement campaign to investigate trace gas transport in convective systems has been deferred to summer 2003 for logistic reasons. Therefore, the high resolution simulations of convective systems and the flash tracer calculations, which were originally planned for 2002, have been deferred to 2003 as well.

The photo-chemical box model simulations were not performed at our institute. Our partner IMK (Institut für Atmosphärenforschung in Garmisch) agreed to take this task.

4. References

Eckhardt, S., H. Wernli, P. James, C. Forster, N. Spichtinger, and A. Stohl, 2002: A 15-year climatology of warm conveyor belts, submitted to Journal of Climate.

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Stohl, A., C. Forster, S. Eckhardt, H. Huntrieser, J. Heland, H. Schlager, H. Aufmhoff, F. Arnold, and O. Cooper, 2002b: A backward modeling study of intercontinental pollution transport using aircraft measurements, Journal of Geophysical Research, in press.

Stohl A., H. Huntrieser, A. Richter, S. Beirle, O. Cooper, S. Eckhardt, C. Forster, P. James, N. Spichtinger, M. Wenig, T. Wagner, J. Burrows, and U. Platt, 2003: Air pollution taking an intercontinental express highway in a bomb, in preparation for Atmos. Chem. Phys.

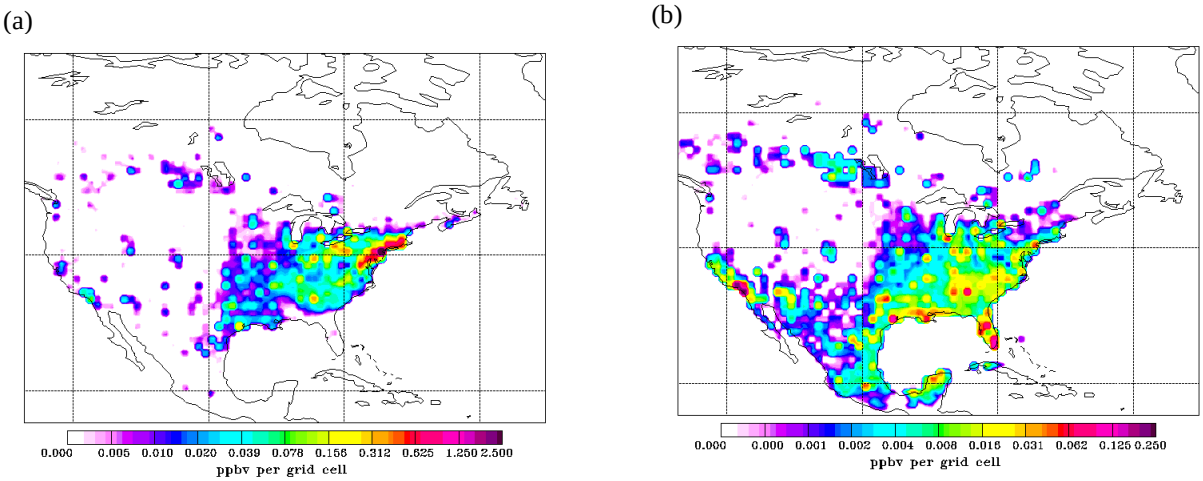

Figure 1: Average source contribution per 0.5° grid box to the total CO mixing ratio measured along the flight track on (a) 19 November 2001 at 11:53 UTC and (b) 27 November 2001 between 14:10 and 14:45 UTC.