

CONTRACE - Convective Transport of Trace Gases into the Middle and Upper Troposphere over Europe

Guest Contribution / EXPORT-E2

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Summary

The objective of the AFO2000-project CONTRACE is to investigate the impact of convective transported air masses on the trace gas composition and budget (mainly NO_x and O₃) in the middle and upper troposphere over Europe. The first airborne field experiment was carried out from southern Germany in November 2001. The DLR research aircraft *Falcon* was equipped with a complex instrumentation to measure NO, NO_y, CO, CO₂, O₃, J(NO₂), acetone, HNO₃, H₂SO₄, SO₂, ions, H₂O₂, formaldehyde, hydrocarbons, J(O¹D) and particles. An extensive set of chemical and meteorological forecast products, including trajectory calculations, was developed and used in combination with satellite images (Meteosat and GOES) to plan the flights. A passive tracer for surface emissions (CO) was used in the forecast models to separate the continental and intercontinental (North America to Europe) transport of polluted air masses. During all CONTRACE flights in the middle and upper troposphere polluted layers with different origin and distinctly enhanced NO, NO_y, CO and CO₂ mixing ratios were found. The European emissions only occasionally reached the middle and upper troposphere. However, more frequently than expected North American emissions were found in the middle and upper troposphere over Europe.

Introduction

Isolated thunderstorms, mesoscale convection in the vicinity of cold fronts, and uplifting of air masses by the large-scale circulation, e.g. warm conveyor belts (WCB), are effective in rapidly transporting trace gases emitted or generated in the boundary layer (BL) into the upper troposphere (UT) (Cotton et al., 1995; Huntrieser et al., 1998). The time scale of this vertical transport is frequently only a few hours and for WCB 1 or 2 days. Once in the UT, the trace gases have substantially longer lifetimes and can be transported over long distances, even between continents. Oxidation products, in particular ozone, can therefore influence the oxidizing capacity and the radiative balance of the UT at super-regional scales. The overall impact of these processes on the distribution and budget of trace gases in the UT over Europe is presently not well known. A major challenge is to quantify the fraction of transported surface-NO_x (=NO+NO₂) emissions found in the UT and to compare it to aircraft- and lightning-produced NO_x (Meijer et al., 2000). Only with this knowledge the importance of the various NO_x sources for the ozone budget in the UT can be obtained and the effect of future source changes be estimated.

Objectives

The main objectives of the CONTRACE-project (2001-2003) are:

- to investigate the relative importance of convective clouds, frontal systems and long-range advection in transporting nitrogen oxides (NO_x), HO_x precursors, and other chemical species into the UT over Europe including variation with season;
- to carry out the first UT peroxy radical measurements (HO_2 , ΣRO_2) over Europe using a novel experimental method (planned for summer 2003);
- to quantify the total convective transport of NO_x and other ozone precursors over Europe;
- to compare the contribution from convection to the trace gas budget of the UT with the contributions by aircraft emissions and lightning;
- to analyse perturbations of the UT by aged pollution layers formed by convection or frontal uplift upwind of Europe;
- to investigate the impact of convectively uplifted trace chemicals on the photochemistry.

Activities

In November 2001 the first CONTRACE field experiment was carried out from the DLR operation site in Oberpfaffenhofen (southern Germany). The DLR research aircraft *Falcon* was used as platform for the in situ measurements in the middle and upper troposphere (MUT). It was equipped with a large variety of instruments (NO , NO_y , CO , CO_2 , O_3 , $\text{J}(\text{NO}_2)$, acetone, HNO_3 , H_2SO_4 , SO_2 , ions, H_2O_2 , formaldehyde, hydrocarbons, $\text{J}(\text{O}^1\text{D})$ and particles) by the research groups from DLR, MPI-Heidelberg and IFU. The planning of the flight missions was supported by an extensive set of chemical and meteorological forecast products (provided by EURAD, MPI-Mainz and TUM), including trajectory calculations (trajectory model FLEXTRA). A passive tracer for surface emissions (CO) was used in the particle-dispersion model FLEXPART (TUM) and in the chemical transport model MATCH (MPI-Mainz) to separate the continental and intercontinental (North America to Europe) transport of polluted air masses. The forecast products were used in combination with current satellite images (Meteosat) to plan the flights that covered a large part of Europe. Further, Meteosat and GOES satellite images were combined to one image (provided by NOAA) for tracking of cloud systems (cyclones) moving from the North American continent to Europe. A special criteria for the selection of warm conveyor belts (ascending region ahead of cold fronts) were used (similar as described in Stohl, 2001). Air mass trajectories that showed a strong ascent (from BL to the MUT) in the past 4 days over the eastern part of the North American continent (most polluted region) were selected. Research flights were performed when these polluted North American air masses were transported to Europe. Further, flight missions were planned when the forecast models indicated frontal uplift of polluted European air masses which was frequently connected with strong outbreaks of cold air masses from the polar region moving quickly over central Europe towards the Mediterranean area. CONTRACE flights were performed on four days (November 14, 19, 22 and 27) with end destinations to Corsica (France), Stockholm (Sweden), Brussels (Belgium) and Budapest (Hungary).

Results

During all CONTRACE flights in the MUT over Europe polluted layers with different origin and distinctly enhanced NO , NO_y , CO and CO_2 mixing ratios were found. The layers can be characterized by calculating deviations from the background profiles (Newell et al., 1999). Typical background composition of the different trace gases in the troposphere during CONTRACE is described next. The vertical ozone profiles (from Oberpfaffenhofen and at the end destinations) showed quite similar behaviour for all flights. The lowest O_3 mixing ratios (20-40 ppbv) were found in the BL. In the free troposphere O_3 was rather constant or slightly increasing with altitude (40-50 ppbv). The flights were performed up to 10 km altitude. The tropopause region was reached only during the last flight (up to 90 ppbv O_3). The background

CO vertical profiles showed a strong maximum in the BL (100-200 ppbv) as expected in winter over Central Europe. In the free troposphere the background CO was quite constant or slightly decreasing with altitude (70-90 ppbv). In comparison the background CO₂ mixing ratio showed a different behaviour with altitude. Only on some days a pronounced maximum was found in the BL (375-385 ppmv). In the free troposphere the background CO₂ showed an increasing mixing ratio with altitude (370-380 ppmv). Because of the pronounced CO (and sometimes CO₂) maximum in the BL, CO and CO₂ were used as tracers for BL air transported into the free troposphere since they both undergo only slow chemistry. Typical NO and NO_y mixing ratios found in the BL were up to 1-2 ppbv and 5-6 ppbv, respectively. In the free troposphere the background mixing ratios varied around 0.05 ppbv NO and 0.1-0.4 ppbv NO_y.

During the CONTRACE field phase the CO tracer forecasts for Europe indicated that the European emissions were only occasionally uplifted to the middle troposphere and seldom to the upper troposphere. During the first flight mission to Corsica (November 14th) enhanced CO mixing ratios (up to 120 ppbv) were measured in the middle troposphere as predicted by the models. Polluted Mediterranean air masses were lifted rapidly ahead of an outbreak of cold air from the north. However, in comparison to the forecasts the polluted air masses (100-120 ppbv CO) even reached altitudes as high as 9 km. The satellite images indicated that thunderstorms close to Corsica could have contributed to this rapid transport to the upper troposphere. The polluted layers in the MUT were also distinct in the CO₂ mixing ratio (380-385 ppmv) and in the NO and NO_y mixing ratios (0.5-1.5 ppbv, up to 3 ppbv, respectively). The rather high NO/NO_y ratio of 0.5 indicated fresh NO sources (possibly production by lightning). On November 22nd an intensive cold front from north west moved over Germany and lifted polluted BL air up to a layer between 2 and 6 km. In this extended polluted layer CO mixing ratios between 120 and 150 ppbv were observed. The highest CO values were combined with very low ozone mixing ratios (20 ppbv) and high CO₂ mixing ratios (380-385 ppmv) indicating a very rapid transport from the BL. The polluted layer was also characterized by high NO_y mixing ratios (1-6 ppbv) and unexpected low NO mixing ratios (below 0.02 ppbv).

The CO tracer forecasts for Europe indicated more frequently than expected North American emissions in the MUT over Europe (extended layers at least once a week). Some of these layers had a horizontal extension of more than 1000 km in one direction. On November 19th North American emissions were predicted to reach parts of Scandinavia (Figure 1). Five days before these air masses were lifted over North America in combination with a warm conveyor belt passing the area around the Great Lakes. The mission flight to Stockholm showed a polluted layer most pronounced between 2 and 5 km altitude (Figure 1). CO mixing ratios between 100 and 160 ppbv, and CO₂ mixing ratios between 375 and 380 ppmv were observed. In comparison to the polluted layers with European origin the North American layer also showed enhanced ozone mixing ratios (5-10 ppbv above background values) indicating ozone production. The NO_y mixing ratio varied between 0.5 and 1 ppbv, and NO was below 0.05 ppbv. A second mission flight to investigate North American emissions over western Europe was started on November 27th. On November 23rd these air masses were lifted very rapidly in connection with a warm conveyor belt (with thunderstorms and tornadoes) over eastern USA. The polluted air masses were observed over Belgium in a rather thin layer around 9 km altitude. CO mixing ratios between 90 and 110 ppbv, and CO₂ mixing ratios around 380 ppmv were observed. Again ozone was slightly enhanced (5-10 ppbv above background values). The NO_y mixing ratio was around 0.5 ppbv and NO around 0.1 ppbv.

Conclusions

The aim of CONTRACE is to investigate the origin, frequency, chemical composition and impact of polluted layers found in the MUT over Europe. During the first field phase in

November 2001 both recently uplifted (European origin) as well as aged polluted layers (from North America) were found during the airborne measurements. Mainly warm conveyor belts (but also thunderstorms) were responsible for the uplift of air masses from the BL. In general the European emissions only reached the lower or middle troposphere, and showed higher mixing ratios of pollutants due to the more recent uplift. However, ozone mixing ratios in these layers were low indicating recent transport from the BL where the lowest ozone mixing ratios were observed. In comparison the horizontal extension of the North American pollution layers (mainly in the MUT) was much larger (over 1000 km) due to fast dispersion of air masses in the frontal zone. The ozone mixing ratios in the polluted air masses from North America were slightly enhanced (5-10 ppbv above background) indicating ozone production. For the first time a combination of chemical and meteorological forecast products were successfully used to direct airborne measurements into polluted layers in the troposphere. Polluted layers from North America have been observed before over Europe, however until now these layers were found more randomly (Arnold et al., 1997; Stohl and Trickl, 1999).

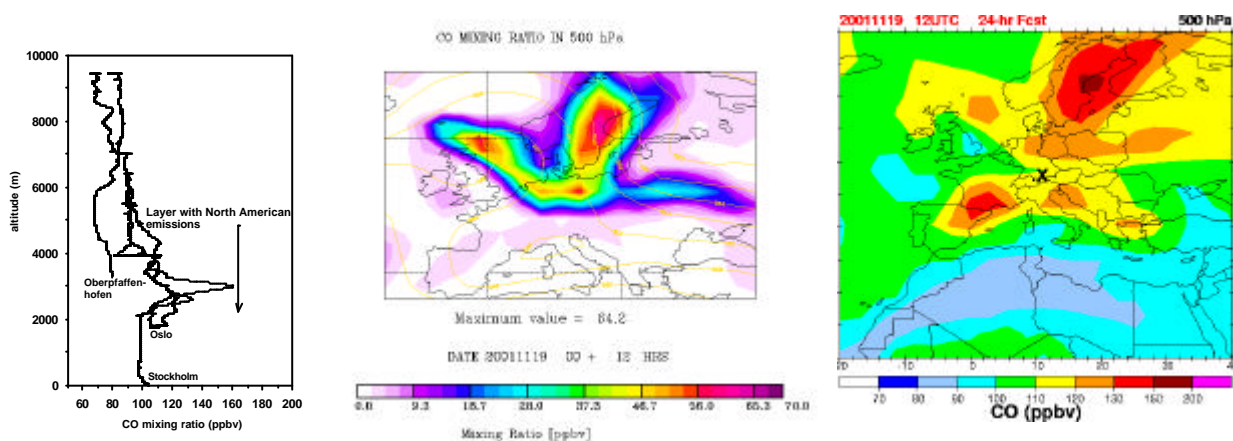


Figure 1: Observed vertical CO distribution on November 19th and predicted CO tracer distribution (TUM, MPI)

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