Intercontinental transport of pollution from North America to Europe: Airborne trace gas measurements over Central and Northern Europe during CONTRACE

A contribution to subproject EXPORT-E2

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Introduction

The objective of the AFO2000-project CONTRACE is to investigate the impact of convective transported (and/or frontal lifting of) air masses from the boundary layer on the trace gas composition and budget (NO_x and O_3) in the middle and upper troposphere over Europe.

The first airborne CONTRACE 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, CO_2, O_3, $J(NO_2)$, acetone, SO_2, ions, H_2O_2, formaldehyde, NMHC, $J(O^1D)$ 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 included in the forecast models to separate the regional and intercontinental (North America to Europe) transport of polluted air masses.

Results

During all CONTRACE flights in the free troposphere polluted layers with different origin (European / North American) and distinctly enhanced trace gas mixing ratios (especially NOy and CO) were successfully observed. The European emissions only occasionally reached the middle and upper troposphere. On 14 November the chemical forecast models indicated lifting of surface emissions in the Mediterranean area ahead of a cold front system that passed over Central and Southern Europe. The airborne measurements show that these emissions were lifted up to 3 km altitude over Corsica. Further, several pollution layers were found in the middle and upper troposphere (4-7 km) over Corsica. The outflow from nearby thunderstorms to west probably caused these enhancements in the CO (120 ppbv), NO (1.5 ppbv) and NO_y (3 ppbv) signals.

For the first time it succeeded to guide the Falcon aircraft were precisely into pollution plumes transported all the way from North America. Until now these plumes had only been observed by coincidence. More frequently than expected significant amounts of North American emissions were found in the troposphere over Europe. During the CONTRACE field phase these kind of pollution events were observed weekly. The forecast models showed how pollution plumes were lifted over Eastern North America, ahead of approaching cold fronts, in so-called warm conveyor belts (WCB) and then rapidly transported to the east. In about 4 days these pollution plumes reached Europe. Three out of four CONTRACE flights were used to probe the pollution plumes from North America (for example case 19 and 22 November).

On 19th November several vertical profiles were flown between Oslo and Stockholm indicating a polluted CO-layer located in the lower and mid troposphere with CO mixing ratios reaching up to 170 ppbv. Surprisingly for the winter season, a positive O_3/CO correlation was observed in the plume and the O_3 mixing ratio increased from 43 to 53 ppbv (ΔO_3 = +10 ppbv). Comparisons with O_3 measurements from a mountain station in Eastern North America, where the passage of the lifted pollution plume was observed, showed enhanced O_3 mixing ratios in the same range (50-60 ppbv). Most likely O_3 was already produced photochemically in the polluted boundary layer over Eastern North America and not in transit over the North Atlantic.

On 22 November a complex chemical weather situation was predicted for Central Europe with lifting of European emissions into the lower troposphere ahead of an approaching cold front and simultaneously, the advection of a pollution plume from Eastern North America in mid tropospheric layers. Both pollution signatures were observed in large detail by the Falcon measurements. Similar CO mixing ratios were observed in both plumes making it difficult to distinguish the two plumes without additional information from other trace gases like NO_y and O_3 (Figure 1). The European pollution plume was characterized by large enhancements in the CO (150 ppbv) and NO_y (6 ppbv) mixing ratios. The NO_y/CO ratio was 0.135 which is a typical value for fresh emissions (age few hours). In comparison the NO_y/CO ratio for the North American pollution plume was 0.010 indicating a tracer age of 4 days. The observed CO and NO_y mixing ratios in this plume were 160 ppbv and 1 ppbv, respectively. The two plumes were also characterized by very different O_3 to CO relationships. In the plume from North America a positive O_3/CO slope was observed similar to the values as described above for the 19 November plume indicating photochemical ozone production. The European plume showed a strong negative O_3/CO relationship with O_3 mixing ratios down to 20 ppbv when the highest CO mixing ratios were observed. The cause of the low O_3 values was the direct upward transport of polluted boundary layer air (in winter low O_3 mixing ratios due to titration by NO). In addition the SO_2-, acetone mixing ratios were especially enhanced in the pollution plumes from North America (2-3 ppbv SO_2 and 5-6 ppbv acetone) in comparison to the observed European plume.

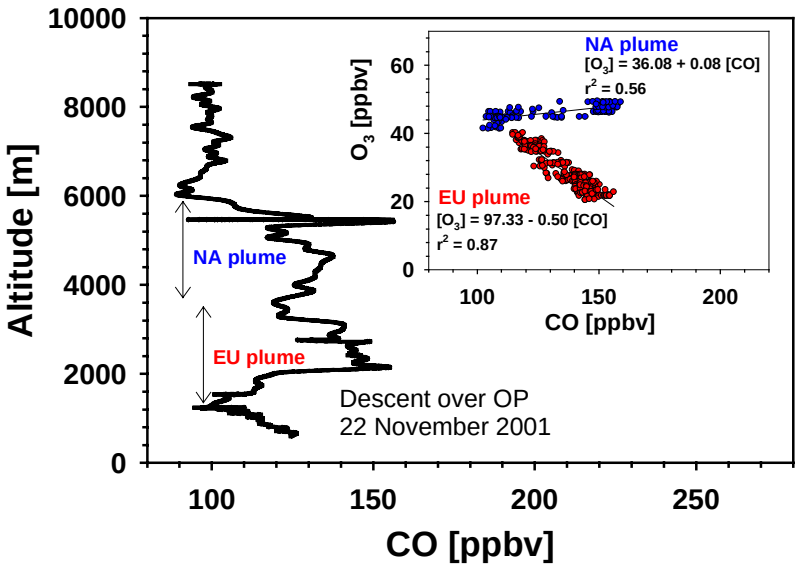

Figure 1. Descent over Oberpfaffenhofen (Germany) on 22 November 2001. Two polluted layers with different origin (European / North American) were penetrated with the Falcon aircraft. In the European (North American) plume a negative (positive) O_3/CO relationship was observed.