

# Analysis of Convective and Long-range Transport of Reactive Nitrogen Species over Europe using Aircraft Observation

A contribution to subproject EXPORT-E2

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## Summary

The vertical transport and the production of  $\text{NO}_x$  by lightning in deep convection were investigated using aircraft measurements. From the observations during KONVEX a mean convective mass flux of  $\text{NO}_x$  into the upper troposphere of 50 and 8 g (N)  $\text{s}^{-1}$  was determined for thunderclouds with and without lightning, respectively. For electrically active thunderclouds 70-80% of the  $\text{NO}_x$  flux out of the anvils was due to  $\text{NO}_x$  produced by lightning. Using statistical data for lightning, the  $\text{NO}_x$  injection into the upper troposphere by electrically active thunderclouds was estimated to 0.5 kg (N)  $\text{km}^{-2} \text{yr}^{-1}$  for the region of Southern Germany. Falcon measurements in the outflow of thunderclouds in central Europe during EULINOX yield a  $\text{NO}_x$  production by lightning on a European scale of 0.03 Tg (N)  $\text{yr}^{-1}$ . Signatures for long-range transport of emissions from North America to Europe were observed during EXPORT and CONTRACE. The observed pollution plumes have been uplifted over Eastern North America in warm conveyor belts and rapidly transported to Europe in the middle and upper troposphere within about 3-5 days. Positive  $\text{O}_3/\text{CO}$  correlations were observed in the North American pollution plumes indicating photochemical  $\text{O}_3$  production.

## Aim of the Research

This project investigated the transport of nitrogen oxides and other pollutants from the planetary boundary layer into the free troposphere and the import/export of pollutants to/from Europe by long-range transport using aircraft observations, air mass trajectory analyses, and satellite imagery. Specific objectives were:

- to quantify the vertical transport of  $\text{NO}_x$  into the upper troposphere over Europe by convective clouds
- to quantify the production of  $\text{NO}_x$  by lightning in thunderclouds
- to study long-range transport of pollutants by warm conveyor belts (WCB).

## Methods used

In situ trace gas measurements from the DLR Falcon were used for the investigations. These data were sampled during the field campaigns KONVEX, EXPORT, and CONTRACE in Central and Eastern Europe. The operation base was DLR Oberpfaffenhofen in Southern Germany. For the

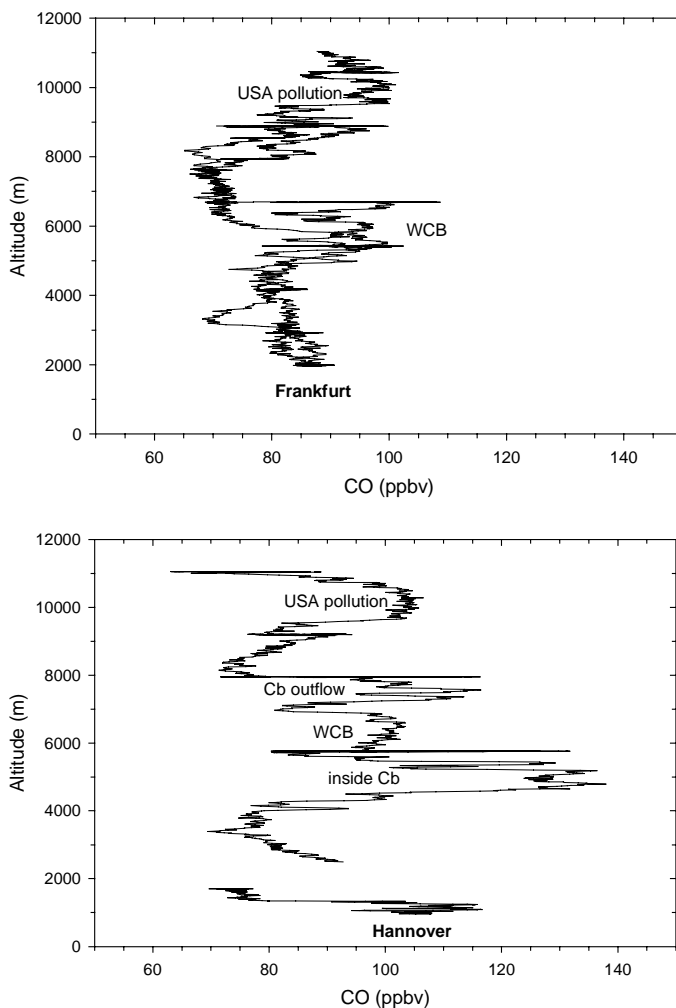
planning of the aircraft flights an extensive set of forecast products were used including air mass trajectories, METEOSAT imagery, and chemical forecast fields. During the campaigns the Falcon was directed into the anvils of convective clouds, electrically active thunderclouds, and pollution layers in the middle and upper troposphere originating from North America.

## Principle Results

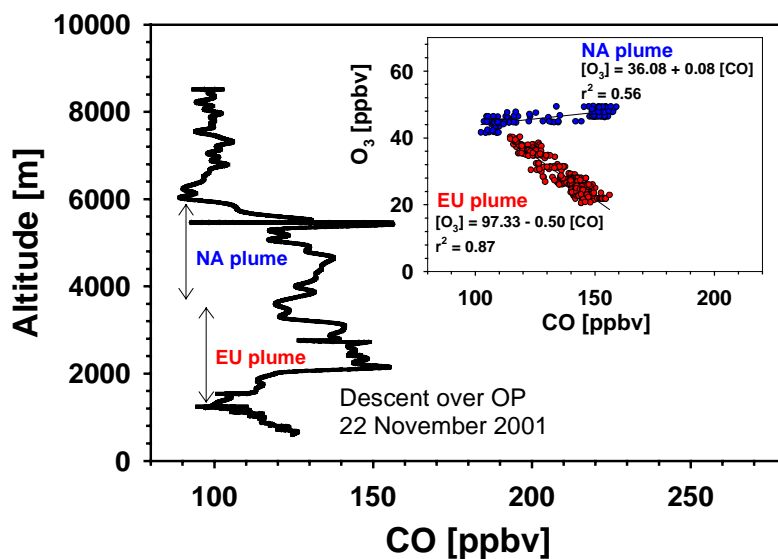
During the KONVEX experiment convective clouds could be penetrated more than 30 times in southern Germany. From the cloud measurements a mean convective mass flux of nitrogen oxides ( $\text{NO}_x$ ) into the upper troposphere of 50 and 8 g (N)  $\text{s}^{-1}$  was determined for thunderclouds with and without lightning, respectively. For the electrically active thunderclouds 70-80% of the  $\text{NO}_x$  flux out of the anvils was due to  $\text{NO}_x$  produced by lightning. Within the typical lifetime of the thunderclouds 40-60 kg (N)  $\text{s}^{-1}$  were transported into the upper troposphere. Using statistical data for lightning, the  $\text{NO}_x$  injection into the upper troposphere by electrically active thunderclouds was estimated to 0.5 kg (N)  $\text{km}^{-2} \text{yr}^{-1}$  for the region of Southern Germany. The observations of lightning  $\text{NO}_x$  in thundercloud anvils are in agreement with earlier measurements performed during the LINOX and EULINOX experiments (Huntrieser *et al.*, 1998, 2002). An estimation of lightning  $\text{NO}_x$  from KONVEX and EULINOX measurements on the European scale amounts to 0.03 Tg (N)  $\text{yr}^{-1}$ .

During the EXPORT campaign numerous elevated pollution layers of different origin were observed in the middle and upper troposphere. Figures 1a and 1b show as an example CO measurements performed on 2 August 2000 over Germany. Depicted are vertical profiles measured over Frankfurt and Hannover. At altitudes between 2 - 6 km layers of enhanced CO mixing ratios are visible in both profiles caused by the large scale lifting of pollutants along an active cold front with a pronounced warm conveyor belt (WCB). The WCB was slanted between Geneva at an altitude of 3-5 km and Hannover (here located at 6-7 km), and consisted mainly of Altostratus and Nimbostratus clouds. In the WCB average CO mixing ratios were 90 and 100 ppbv, i.e. about 10-20 ppbv above the CO background mixing ratios. At the most northern site (Hannover) further pollution layers were observed caused by thunderstorms that were embedded in the cold front. The strongest CO enhancement was observed inside a thundercloud that was penetrated between 4.5 and 6 km with maximum mixing ratios of 130 ppbv. Similar CO mixing ratios were found in the boundary layer below the thunderstorm indicating that the vertical transport of polluted boundary layer air was fast and almost undiluted. In addition, a pollution layer was measured in the uppermost troposphere originating from North America.

During CONTRACE pollution plumes were frequently observed in the middle and upper troposphere that were uplifted over Eastern North America in warm conveyor belts associated with cold fronts. As an example Figure 2 shows measurements from a Falcon flight on 22 November 2001 where elevated pollution plumes from North America and European emission sources were present. Similar CO mixing ratios were observed in both plumes. The European pollution plume was characterised by large enhancements in the CO (150 ppbv) and  $\text{NO}_y$  (6 ppbv) mixing ratios. The  $\text{NO}_y/\text{CO}$  ratio was 0.14 which is a typical value for fresh emissions aged only a few hours (according to tracer simulations). In comparison the  $\text{NO}_y/\text{CO}$  ratio for the North American pollution plume was 0.01 indicating an age of about 4 days. The observed CO and  $\text{NO}_y$  mixing ratios in this plume were 160 ppbv and 1 ppbv, respectively. The two plumes were also characterised by very different  $\text{O}_3/\text{CO}$  correlations. In the North America pollution plume a positive  $\text{O}_3/\text{CO}$  correlation was observed indicating photochemical ozone production. The European pollution plume showed a strong negative  $\text{O}_3/\text{CO}$  correlation. The low  $\text{O}_3$  values at high CO mixing ratios are probably due to titration of  $\text{O}_3$  by NO co-emitted with CO.



**Figure 1:** Vertical CO profiles measured with the DLR Falcon on 2 August 2000 over Frankfurt and Hannover. A Warm Conveyor Belt (WCB) with enhanced CO mixing ratios was penetrated in the mid troposphere. In the uppermost troposphere a layer with pollution from North America was measured. At Hannover also thunderstorms (Cb) embedded in the WCB were penetrated.



**Figure 2.** Aircraft profile measurement over Oberpfaffenhofen (OP) on 22 November 2001. Two polluted layers with different origin (European / North American) were penetrated with the Falcon. In the European and North American pollution plume a negative and positive O<sub>3</sub>/CO correlation was observed, respectively.

## Main Conclusions

The vertical transport of NO<sub>x</sub> by deep convection was investigated in detail for a region in central Europe during KONVEX. It was possible to infer from the Falcon in situ measurements a mean convective mass flux of NO<sub>x</sub> into the upper troposphere of 50 and 8 g (N) s<sup>-1</sup> for thunderclouds with and without lightning, respectively. Based on the KONVEX and earlier EULINOX data and statistical data for lightning, the NO<sub>x</sub> injection into the upper troposphere by electrically active thunderclouds was estimated to 0.5 kg (N) km<sup>-2</sup> yr<sup>-1</sup> for the region of Southern Germany and 0.03 Tg (N) yr<sup>-1</sup> for the European scale. For comparison, aircraft emissions over Europe amount to 0.1 Tg (N) yr<sup>-1</sup>.

During EXPORT and CONTRACE the DLR Falcon was successfully guided into pollution plumes originating from European and North American pollution source regions using air mass trajectory forecasts with a passive tracer for surface emissions (CO). North American emissions were more frequently found in the middle and upper troposphere over Europe than expected. The European pollution plumes were mostly observed in the lower troposphere and reached only occasionally the middle and upper troposphere.

## Future Aims

It is planned to participate in the INTEX (Intercontinental Chemical Transport Experiment) field campaign in summer 2004 with Falcon in situ measurements of chemical species and aerosols. INTEX aims for an understanding and quantification of the exchange of trace species between continents in the northern hemisphere.

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## References

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