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Regional nitric oxide enhancements in the North Atlantic flight corridor observed and modeled during POLINAT 2 – a case study

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Abstract. In situ measurements of nitrogen oxides and other trace chemicals were performed aboard the DLR Falcon in September and October 1997 during POLINAT 2 over the eastern North Atlantic in areas with predicted high impact of aircraft emissions to search for flight corridor effects. During survey flights in the upper troposphere from the centre of the flight corridor to regions south of or below the major transatlantic aircraft routes, large-scale enhancements in mixing ratios of NO of about 50 to 150 pptv were observed in corridor areas compared to the NO abundance measured outside the corridor. Using simultaneous tracer measurements, back trajectory analyses for the air masses sampled, the actual distribution of the North Atlantic air traffic, and comparisons of observed and predicted NO distributions from a regional model of simulations including or excluding aircraft emissions, these enhancements were attributed mainly to aircraft NO_x.

Introduction

Subsonic aircraft release emissions into the upper troposphere and lowermost stratosphere during cruise. Nitrogen oxide (NO_x = NO + NO_2) emissions are of particular interest due to their potential to impact the ozone abundance near the tropopause and thereby the climate. A number of model studies suggest that NO_x emissions from aircraft may have increased the NO_x abundance near the tropopause at northern mid-latitudes by up to 100 % [e.g. Brasseur et al., 1996; Flatoy and Hov, 1996]. Strongest perturbations by aircraft NO_x are expected in heavily travelled flight corridors over the North Atlantic and the North American and European continent. Small-scale signatures of relatively fresh aircraft NO_x have been observed by recent airborne measurements in regions with dense air traffic [Zheng et al., 1994; Schlager et al, 1997, Schumann et al., 1998]. Large-scale corridor effects by aircraft NO_x are difficult to detect due to the influence of other NO_x sources on the NO_x abundance near the tropopause including surface emissions, lightning, and input from the stratosphere. Recent airborne observations downstream of an area over the eastern USA associated with heavy air traffic [Witte et al., 1997] and in a stagnant anticyclone located in the North Atlantic flight corridor [Schlager et al., 1996] indicate that regional enhancements in NO due to aircraft emissions may occur.

During the POLINAT 2 (Pollution from Aircraft Emissions in the North Atlantic Flight Corridor) project [Schumann et al., 1999; Singh et al., 1999] focused aircraft measurements were performed over the eastern North Atlantic to search for large-scale enhancements of air-traffic related trace species. Here we present results of two flights conducted in regions with expected high impact of aircraft emissions. These airborne observations to study large-scale flight corridor effects were planned using a multitude of forecast products and other information including the location of the Organized Track System (OTS) for transatlantic air traffic, air mass back-trajectory forecasts, and forecasts of meteorological and chemical fields. In comparing the data obtained to results of a regional chemistry transport model we examine accumulations of aircraft NO_x on horizontal scales of several 100 kilometres.

Measurements and Regional Model

The POLINAT 2 mission was performed in September/October 1997 out of Shannon, Ireland. Simultaneous measurements of trace gases including NO, NO_2, O_3, and CO, and meteorological parameters were made over the eastern North Atlantic using the DLR Falcon. The flight patterns for observations of large-scale corridor effects included constant altitude survey flights from the centre of the OTS to regions south of the flight corridor at flight levels with most dense air traffic (e.g. 35,000 feet = FL350 ~10.7 km) and flight levels located just below the altitudes of transatlantic air traffic (e.g. FL290). We present results of two long-range flights from Shannon to the Azores and Shannon to Tenerife performed on 21 September and 14 October 1997, respectively, during daytime.

Volume mixing ratios of NO_x were measured by chemiluminescence technique including a photolysis cell for NO_2 detection [Ziereis et al., 1999]. O_3 and CO were measured by UV-absorption [Schlager et al., 1997] and VUV fluorescence [Gerbig et al., 1996], respectively. The nominal accuracy of the NO, NO_2, O_3, and CO measurements are 10%, 20%, 5%, and 10%, respectively.

Forecasts and analyses of chemical fields over the North Atlantic were calculated with the NILU three-dimensional regional chemistry transport model (NILU-CTM) described in Flatoy and Hov [1996]. The chemical scheme of the model includes 51 species. The horizontal grid resolution of the large (northern hemisphere) and small (Europe) domains of the

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model is 150 and 50 km, respectively, including 10 and 18 unequally spaced layers, respectively. The meteorological input to the NILU-CTM consists of meteorological forecasts from a weather prediction model employed on the CTM domains and using ECMWF (European Centre for Medium-range Weather Forecasts) analysis and forecasts as input.

To distinguish the role of aircraft NOx emissions in the determination of the NOx distribution near the tropopause from contributions of other NOx sources, model runs were performed with aircraft NOx emissions switched on and off, respectively. Details on the model set-up and products of the chemical forecasts used for flight planning are given by Flatoy et al. [1999].

September 21 Flight

Figure 1 depicts the flight path of September 21, 1997 from Shannon to the Azores together with the location of the OTS routes over the North Atlantic for the mission day and two preceding days. Also included are 3-day backward trajectories starting from points along the flight track at a constant pressure level of 239 hPa (FL350) for every 2 minutes of flight time. These trajectories are based on ECMWF winds (analyses) for the measuring period. Air mass travel times of one day are indicated by different colours. The flight was designed to sample air masses in and out of the flight corridor. As shown by the back-trajectories, the air parcels sampled experienced no vertical displacement 3 days prior to the measurements. Air parcels probed north of 50°N spent about 2 - 3 days in the OTS region whereas air parcels measured south of 50°N originate from the mid-Atlantic with very low probability of air traffic encounters.

Figure 2 shows calculated fields of total NO (top panel) and aircraft NO (bottom panel) at a pressure level of 239 hPa (FL350) for the measurement area and period. Given are results of model runs using ECMWF analyses data which confirmed the corresponding chemical forecasts used for the planning of this flight. West of Ireland a pronounced region with elevated concentrations of NO can be noticed. Here total NO is predicted to be mainly due to aircraft NO.

Observed concentrations of NO, O_3, and CO along the Falcon track are shown in Figure 3 together with the pressure altitude of the aircraft. The O_3 trace indicate that the entire flight took place below the tropopause, except a small section around 52°N where the Falcon flew close to the tropopause. According to the NO_2 measurements (not shown), nitrogen oxides were mainly made up of NO with an observed NO/NOx ratio in the upper troposphere of about 0.7. The median NO/NOy ratio measured during the POLINAT 2 flights around the tropopause amounts to 0.2. South of 50°N, the observed NO concentrations are mostly below about 50 pptv with the exception of a few NO peaks representing most probably aged aircraft plumes [Schlager et al., 1997]. North of 50°N, in the region with predicted accumulation of aircraft emissions, however, NO concentrations are higher by a factor of about 2 to 4 ranging between 100 and 200 pptv. In addition, a number of aged aircraft plumes are also visible in this flight section with NO concentrations up to 2 ppbv. In contrast, observed CO concentrations (tracer for surface emissions) are not increasing north of 50°N.

For comparison, calculated NO mixing ratios interpolated along the Falcon track from the NILU-CTM simulations are included in Figure 3. Given are results from two model runs: full model including all NO_x sources (dashed trace) and model run with aircraft NO_x switched off (dotted trace). According to

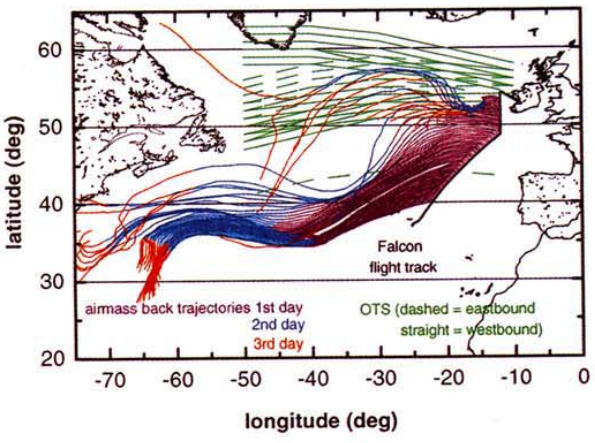

Figure 1. Flight track of Sept. 21, 1997. Also shown are the aircraft routes of the Organized Track System over the North Atlantic of Sept. 19, 20, and 21, and 3-day backward trajectories from the flight track.

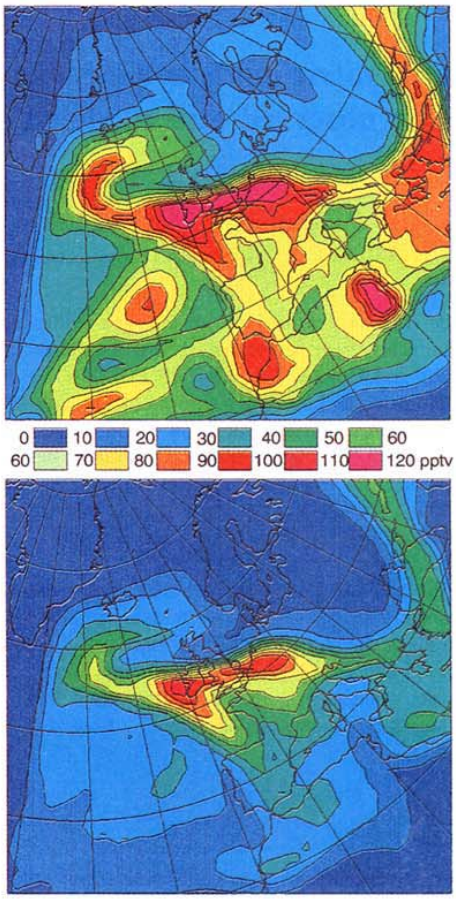

Figure 2. NO mixing ratios at 239 hPa for Sept. 21, 9.00 UTC derived from NILU-CTM runs (upper panel: total NO, lower panel: aircraft NO).

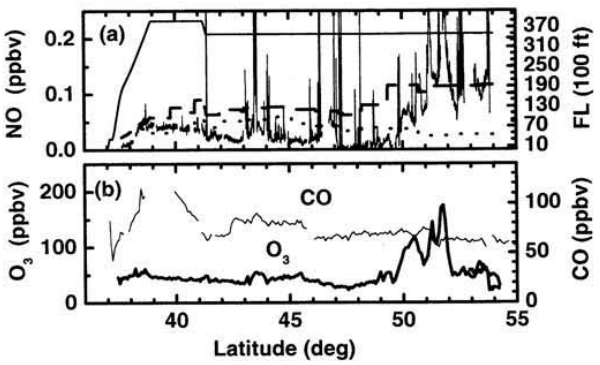

Figure 3. Latitudinal distribution of Falcon measurements from Sept., 21 flight. (a) NO and pressure altitude (solid trace), (b) O_3 and CO. In panel (a) calculated NO mixing ratios from NILU-CTM are included for comparison (dashed trace: full model, dotted trace: aircraft emissions switched off).

the model results, 80% of total NO is due to aircraft emissions in the region north of 50°N. Here total NO derived from the model agree well with the observations. South of 50°N the measured NO abundance is much less affected by aircraft emissions and total NO derived from the model differ from the observations by about a factor of two.

October 14 Flights

Figure 4 depicts the flight tracks of October 14, 1997 from Shannon to Tenerife and back at a constant pressure level of 239 hPa (FL350 with the maximum load of North Atlantic air traffic) and 315 hPa (FL290, below North Atlantic traffic routes). Again, the location of the OTS routes of the mission day and two preceding days is shown together with 3-day air mass backward trajectories from points along the Falcon track at FL350.

Figure 5 shows calculated fields of total NO (top panel) and aircraft NO (bottom panel) at a pressure level of 239 hPa for October 14. A large-scale source signature of aircraft emissions was predicted along the entire measuring track at the corridor flight level with increasing impact of aircraft emissions from the south to the north.

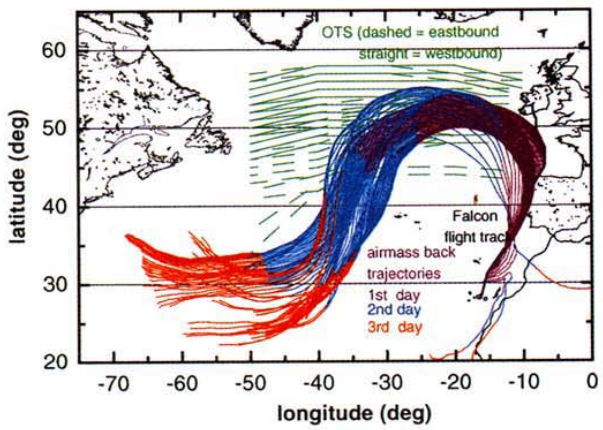

Figure 4. Flight tracks of Oct. 14, 1997. Also shown is the location of the OTS routes of Oct. 12, 13, and 14, 1997 and air parcel backward trajectories from the flight track at FL350.

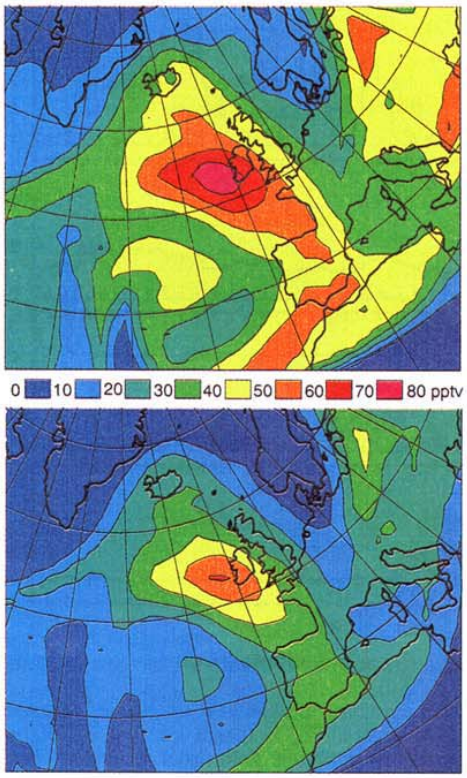

Figure 5. NO mixing ratios at 239 hPa for Oct., 14, 12.00 UTC derived from NILU-CTM runs (upper panel: total NO, lower panel: aircraft NO).

The latitudinal distribution of the NO, O_3, and CO concentrations observed at the two flight levels are depicted in Figure 6. Both flight tracks were located below the tropopause as indicated by the ozone measurements. Again, NO_x was mainly made up by NO. South of 37°N at both flight levels moderately enhanced NO concentrations of about 50 - 100 pptv were measured. Back trajectories for this region indicate that the air masses probed here had 5 days ahead resided over the Caribbean Sea with some expected impact from convective and lightning activity. At latitudes north of 37°N marked differences in the NO abundance were found for the two measuring flight levels (100 - 150 pptv at the corridor flight level and only about 20 pptv at the flight level below the corridor) whereas CO concentrations were very similar. Back-trajectories for the region north of 37°N reveal that the probed air masses of both flight levels originate from the marine boundary layer of the mid-Atlantic, a source region with expected very low NO_x abundance, and had been uplifted and advected over the eastern North Atlantic. However, the air masses probed at FL350 had been uplifted to altitudes of the flight corridor whereas the air masses probed at FL290 resided always below the corridor flight levels. At FL350 in the OTS region north of 45°N, a large number of fresh and aged aircraft plumes were detected. The measured CO trace shows no signature of polluted continental boundary layer air for both flight levels.

For comparison, interpolated NO concentrations of NILU-CTM from the full model run (dashed trace) and a model run with no aircraft emissions (dotted trace) are included in Figure 6. According to the model results, about 50% and 70% of total

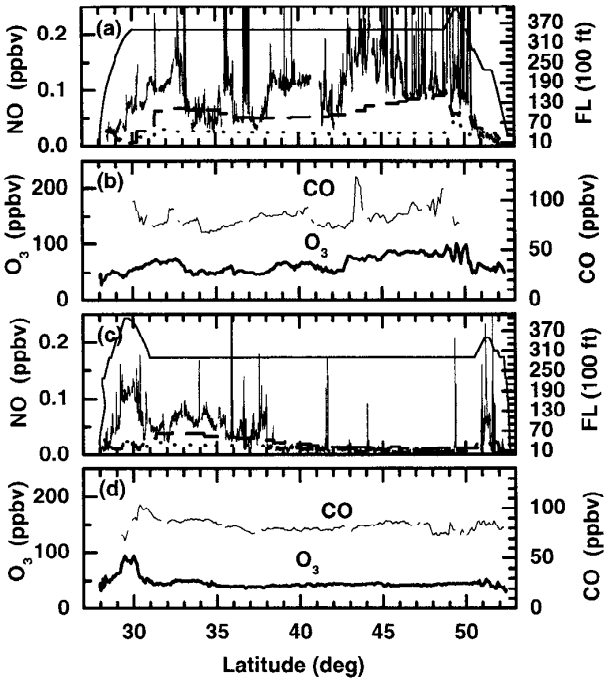

Figure 6. Latitudinal distribution of Falcon measurements from Oct. 14 flight at FL350 (panel a, b) and FL290 (panel c, d). Parameters as in Figure 3. In panel (a) and (c) calculated NO mixing ratios from NILU-CTM are included for comparison (dashed trace: full model, dotted trace: aircraft emissions switched off).

NO is due to aircraft emissions at FL350 in the southern and northern part of the measuring area, respectively. At the flight level below the corridor (FL290) the impact of aircraft emissions is negligible with exception of the most southern part. Near Shannon the Falcon climbed from FL290 into the flight corridor (Figure 6, panel c, north of 50°N). In doing so, the measured NO concentration strongly increased compared to the values below the corridor in agreement with an predicted increase of the NO abundance due to aircraft NO while the CO concentrations remained constant.. In general, model results for total NO agree fairly with observations in the northern part with the largest impact of the air traffic emissions. In other parts of the investigation area, model NO concentrations are smaller than the measured ones by about a factor of two.

Conclusions

During long-range survey flights in the upper troposphere over the eastern North Atlantic large-scale enhancements in NO concentrations were found in air masses which spent 2 to 3 days in the region of the North Atlantic flight corridor. Back-trajectory calculations and tracer (O_3, CO) measurements indicate that the NO enhancements observed were not due to stratospheric input or fast vertical transport of NO_x rich air masses from the boundary layer. Comparison of the observations to simulations with a regional chemistry transport model including model runs with aircraft emissions switched on and off indicate that these regional NO enhancements of about 50 - 150 pptv were mainly due to air traffic emissions. Measurements and model results fairly agree for regions with high impact of aircraft emissions but differ by a factor of about 2 to 3 in regions with minor impact of air traffic. This is likely to be caused by the uncertain contributions from lightning induced NOx and the less well characterized upward convective transport of surface NOx emissions in the model.

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