

Intercontinental air pollution transport from North America to Europe: Experimental evidence from airborne measurements and surface observations

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[1] During the airborne CONTRACE field experiment carried out in November 2001 a number of polluted layers of North American (NA) origin were observed in the free troposphere over Europe. For the first time, forecasts from a Lagrangian particle dispersion model were used to predict the NA pollution events and to direct a research aircraft very precisely into these polluted layers above Europe. Two of the NA pollution events are investigated here: one in detail (case 19 November) and a second more briefly (case 22 November). An exceptional result was that the first pollution plume could be traced with the model and trace gas measurements (airborne and surface) for a period of one week, from the source region over the eastern United States to its decay over the Alps. On 14–15 November a warm conveyor belt lifted the leading edge of the pollution plume over the eastern United States to the mid troposphere where it remained during the transport over the Atlantic. On 19 November the plume was intersected with the research aircraft over Scandinavia at an altitude between 2 and 4 km. Elevated CO (170), O₃ (53), NO_y (1.1), acetone (5.0), and SO₂ (2.6) mixing ratios (nmol mol⁻¹) were measured. A positive O₃-CO correlation was observed in the plume. The observations indicate that the enhanced levels of ozone were already produced near the source region over the eastern United States and not during the transit. In the next days one branch of the plume then turned to the south and descended to ground level over the Alpine region. Elevated O₃ (54 nmol mol⁻¹) and CO (168 nmol mol⁻¹) were observed at the mountain site Zugspitze (southern Germany) during two days. At the Arosa Alpine site in Switzerland the highest daily ozone means of November 2001 were observed during this event.

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1. Introduction

[2] Public and scientific interest in intercontinental air pollution transport has grown tremendously over the last

decade. In 1998, first satellite observations from space showing the transport of a huge yellow dust cloud from the Gobi desert to Washington State across the entire length of the Pacific Ocean were published [Wilkening *et al.*, 2000; Husar *et al.*, 2001; McKendry *et al.*, 2001]. Recently it was reported that a Chinese dust plume reached the French Alps after 10 days of transport across the Pacific and Atlantic Ocean [Grousset *et al.*, 2003]. Even hemispheric-scale transport has been observed now. In summer 2003 a huge smoke cloud from forest fires over Russia was transported around the world in only 17 days [Damoah *et al.*, 2004].

[3] In addition to the long-range transport of mineral dust and forest fire smoke, events with anthropogenic pollution transported from one continent to another may occasionally influence regional air quality of a downwind continent [McKendry *et al.*, 2001]. Since a few years global monitoring from space of anthropogenic trace gases like CO, CH₄, O₃, NO₂ and other minor trace gas constituents is possible with the MOPITT (Measurements Of Pollution In The Troposphere), the GOME (The Global Ozone Monitoring Experiment) and the SCIAMACHY (Scanning Imaging

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Absorption Spectrometer for Atmospheric CHartography) instruments. Recently, the first satellite observation of intercontinental transport of a nitrogen oxide pollution plume from South Africa to Australia was reported by *Wenig et al.* [2003]. However, in many cases the monitoring of single pollution events is still difficult because of the patchy satellite images.

[4] For environmental policy makers these pollution events can nevertheless be of great concern, since they may lead to more frequent noncompliance with national air quality standards [*Jacob et al.*, 1999; *Creilson et al.*, 2003]. It is also important to take pollution trends from upwind continents into account for defining official air quality standards, i.e. to consider regional pollution in a global context [*Collins et al.*, 2000; *Prather et al.*, 2003].

[5] Most research up to now on intercontinental air pollution transport has focused on the impact of Asian emissions on air quality in North America. A number of model studies are available at present [*Berntsen et al.*, 1999; *Yienger et al.*, 2000; *Wild and Akimoto*, 2001; *Li et al.*, 2002], however, only few reports on evidence from experimental observations existed up to recently [*Jaffe et al.*, 1999; *Bailey et al.*, 2000; *Jaffe et al.*, 2003]. Results from a field study in spring 2002 have now broadened the knowledge on the transpacific pollution transport [*Cooper et al.*, 2004a, 2004b; *de Gouw et al.*, 2004; *Goldstein et al.*, 2004; *Nowak et al.*, 2004; *Price et al.*, 2004].

[6] Less attention has been paid to the impact of North American emissions on European air quality [*Pochanart et al.*, 2001; *Creilson et al.*, 2003]. Experimental evidence for pollution transport from North America to Europe is very sparse. A recent study of long-term ozonesonde data from Hohenpeißenberg (Germany) and Payerne (Switzerland) indicates that intercontinental pollution transport has a significant influence on O₃ mixing ratios in the lower troposphere and the boundary layer over Europe [*Naja et al.*, 2003]. In contrast, *Derwent et al.* [1998] pointed out that pollutants from North America (single events) are hardly ever observed at ground sites in Europe. One “textbook” example of long-range air pollution transport from North America to Germany observed with an O₃ lidar has been published by *Stohl and Trickl* [1999] and *Trickl et al.* [2003]. In contrast, the export of North American pollution plumes into the North Atlantic Ocean has been investigated in great detail during, e.g., the North Atlantic Regional Experiment (NARE) [*Fehsenfeld et al.*, 1996; *Wild et al.*, 1996]. For more information on previous experimental results the reader is referred to review articles on intercontinental air pollution export from North America to the eastern North Atlantic and to Europe by *Stohl and Trickl* [2001], *Stohl et al.* [2003a], and *Huntrieser and Schlager* [2004].

[7] Lofting due to a sea breeze front is a common transport process for the export of North American pollutants into the North Atlantic Ocean [*Angevine et al.*, 1996]. Thereby the polluted air mass becomes stably stratified and spreads out in a thin layer 1–2 km above the surface. Depending on the meteorological situation (e.g., existence of an Icelandic low pressure system) this low-level pollution layer can be transported all the way to Europe [*Li et al.*, 2002]. However, the major processes responsible for vertical uplifting of polluted air masses to higher altitudes over

the United States are synoptic-scale warm conveyor belts (WCBs) associated with frontal systems [*Moody et al.*, 1996; *Stohl and Trickl*, 1999; *Parrish et al.*, 2000; *Cooper et al.*, 2001; *Stohl*, 2001; *Cooper et al.*, 2002; *Eckhardt et al.*, 2004], and meso-scale deep convection [*Dickerson et al.*, 1987; *Milne et al.*, 2000]. When pollutants are transported to the mid or upper troposphere, they can easily enter into the jet stream and be transported rapidly to Europe. An airstream climatology by *Stohl* [2001] showed that Europe frequently receives the outflow from WCBs that originate along the eastern seaboard of North America.

[8] In this paper we investigate an event of intercontinental air pollution transport from North America to Europe observed on 19 November 2001 during the CONTRACE (Convective Transport of Trace Gases into the Middle and Upper Troposphere over Europe: Budget and Impact on Chemistry) field experiment. The pollution event was not seen in any available satellite data for the flight planning, but predicted by the FLEXPART Lagrangian particle dispersion model [*Stohl et al.*, 2003a] and by the MATCH-MPIC global model of atmospheric transport and chemistry [*Lawrence et al.*, 2003]. Both models use CO as a pollution tracer. CO is an ideal tracer for long-distance transport of pollution, due to its long photochemical lifetime of several months [*Seinfeld and Pandis*, 1998]. For the first time a research aircraft, in this case the DLR *Falcon*, was directed into a pollution plume transported all the way from North America to Europe. The pollution plume is tracked from its uplift over the eastern United States to its decay over the Alps. We focus on the impact of this plume on the trace gas distribution in the lower free troposphere and boundary layer over Europe.

[9] The paper is structured as follows: Important factors affecting the ozone concentration during the transatlantic transport and its impact on European surface ozone concentrations are summarized in section 2. In section 3 the CONTRACE field experiment, the airborne instrumentation and the FLEXPART model are introduced. The model predictions and the research flight on 19 November 2001 are described in section 4. The meteorological and chemical weather situation over the United States and Europe in November 2001 is presented in sections 5 and 6, respectively. Results from a second research flight through a North American pollution plume on 22 November 2001 are briefly described in section 7. In sections 8 and 9 we discuss and summarize our findings.

2. Factors Affecting Ozone Concentration During Transatlantic Transport and Its Impact on European Surface Ozone Concentrations

[10] Ozone (O₃) plays an important role in tropospheric photochemistry since it is the primary source of the hydroxyl radical (OH), a key tropospheric oxidant. Hydroxyl radicals are produced when O₃ is photolyzed by UV radiation in the presence of water vapor [e.g., *Thompson*, 1992]. Due to the long photochemical lifetime of O₃ in winter, on the order of ~200 days [*Liu et al.*, 1987], it can also be transported over long distances. *Fishman et al.* [1980] published one of the first reports on the simultaneous presence of O₃ and CO bands in the troposphere. They suggested that O₃ was photochemically produced in these polluted layers by the

oxidation of CO and hydrocarbons in the presence of nitrogen oxides.

[11] Important factors affecting the O₃ concentration in polluted air masses during the transport across the North Atlantic are discussed next. One of the key questions is whether the initial O₃ concentration increases or decreases during the transatlantic transport. The transport altitude is critical for the ozone production/destruction [Wild *et al.*, 1996; Reeves *et al.*, 2002]. In the upper troposphere photochemical O₃ production seems to dominate. For air parcels traveling in the boundary layer and mid troposphere, net O₃ destruction is more predominant. Apart from the transport altitude, the initial concentrations of O₃ precursors also affect the O₃ production potential [Wild *et al.*, 1996]. The photochemical O₃ production is mainly controlled by the NO_x (=NO + NO₂) concentration. Frequently it was suggested that the high O₃ concentrations observed downwind of North America were produced photochemically prior to export from the North American boundary layer [Schultz *et al.*, 1998; Stohl and Trickl, 2001; Li *et al.*, 2002]. The dynamical processes (initial conditions in the boundary layer and the advection field) seem to dominate over the photochemical production/destruction of O₃ during in-transit [Flatøy *et al.*, 1996]. Dilution with background air represents another important mechanism which can dominate over photochemical O₃ production for an air parcel traveling across the North Atlantic [Wild *et al.*, 1996].

[12] Furthermore, Wild and Akimoto [2001] performed simulations with a 3-D chemical transport model to investigate the impact of O₃ precursors from anthropogenic sources on downstream continents. The O₃ export from North America to Europe was the most pronounced of all intercontinental transport scenarios due to the rapid and direct transport over the relatively short distance between these two continents. However, there is a lack of studies showing that these polluted air masses of North American origin actually reach European surface sites. Calculations with the Lagrangian particle dispersion model FLEXPART by Stohl *et al.* [2002a], which include CO tracers from different continents, can give an explanation for this. In spring and early summer the polluted North American layers most frequently enter Europe at 5–8 km altitude and north of 60°N. Thereafter, the North American pollution generally moves southward and downward toward the surface south of the Pyrenees and Alps. This descent of air masses containing high concentrations of O₃ and its precursors in combination with warm and sunny weather seems to be very effective for additional O₃ production. The Mediterranean region is known to be one of the regions in Europe with the highest O₃ mixing ratios in summer [Millán *et al.*, 1997], which are currently close to levels being harmful to vegetation [Fumagalli *et al.*, 2001]. It has been suggested that intercontinental pollution transport may partly contribute to this Mediterranean O₃ maximum [Lelieveld *et al.*, 2002; Stohl *et al.*, 2002a].

[13] The impact of North American anthropogenic emissions on surface O₃ in Europe has also been studied by Li *et al.* [2002]. They performed simulations with a global 3-D model of tropospheric chemistry (GEOS-CHEM) for a period of 5 years (1993–1997). Especially during “events” of pollution transport from North America in

summer the surface O₃ concentration over Europe increased significantly (5–10 nmol mol⁻¹). On average the transport of North American pollution caused an O₃ increase of 2–4 nmol mol⁻¹ over continental Europe and of 5 nmol mol⁻¹ at Mace Head. Li *et al.* [2002] also found that 20% of the violations of the European Council O₃ standard (55 nmol mol⁻¹, 8-hour average) in summer 1997 were caused by O₃ import from North America.

3. CONTRACE Field Experiment

3.1. Observations of North American Pollution Plumes

[14] One of the objectives of the CONTRACE project was to investigate the origin of polluted layers found in the free troposphere over Europe and their impact on chemistry. More frequently than expected from earlier studies, strong pollution plumes from North America were predicted to pass over Europe with the FLEXPART Lagrangian particle dispersion model, which was used for the flight planning during the CONTRACE field phase. The pollution plumes were all uplifted over North America in warm conveyor belts ahead of cold fronts [Carlson, 1980; Bethan *et al.*, 1998; Cooper *et al.*, 2001, 2002]. During the 3-week field phase in November 2001 four events of intercontinental air pollution transport from North America to Europe were predicted by the FLEXPART model and also successfully observed (see publications listed below). The first event (11 November 2001) was detected in GOME (Global Ozone Monitoring Experiment) NO₂ data and has been described by Stohl *et al.* [2003b]. They called the transport pathway an intercontinental express highway because of the rapid transatlantic transport (1–2 days) involving a meteorological bomb. The Falcon aircraft was not yet available for measurements of this first event. The second pollution event approaching Europe on 19 November was observed in detail by the Falcon aircraft on a flight from Germany to Scandinavia (Figure 1). Model studies for this event have already been published by Lawrence *et al.* [2003] and Stohl *et al.* [2003a], where the focus was on the transatlantic transport. In this paper we study the dispersion of the pollution plume over the European continent, and report on results from airborne measurements and surface observations. A third pollution event passed over Germany on 22 November, which is also briefly described in this paper. The last pollution event on 27 November was penetrated by the Falcon aircraft over Belgium and has been described elsewhere [Stohl *et al.*, 2003a].

3.2. Airborne Instrumentation

[15] The airborne measurements during CONTRACE were performed with the DLR Falcon research aircraft, which operated mainly in the mid and upper troposphere (5–10 km). An overview of the chemical instrumentation installed in the Falcon is presented in Table 1. For a more detailed description of the instrumentation, which has been used in many previous airborne field experiments, see references listed in Table 1. Details are also given in more recent papers by Ziereis *et al.* [2000] and Huntrieser *et al.* [2002]. Position, altitude, temperature, humidity, pressure, and wind were measured with the standard Falcon meteorological measurement system. All Falcon altitude values

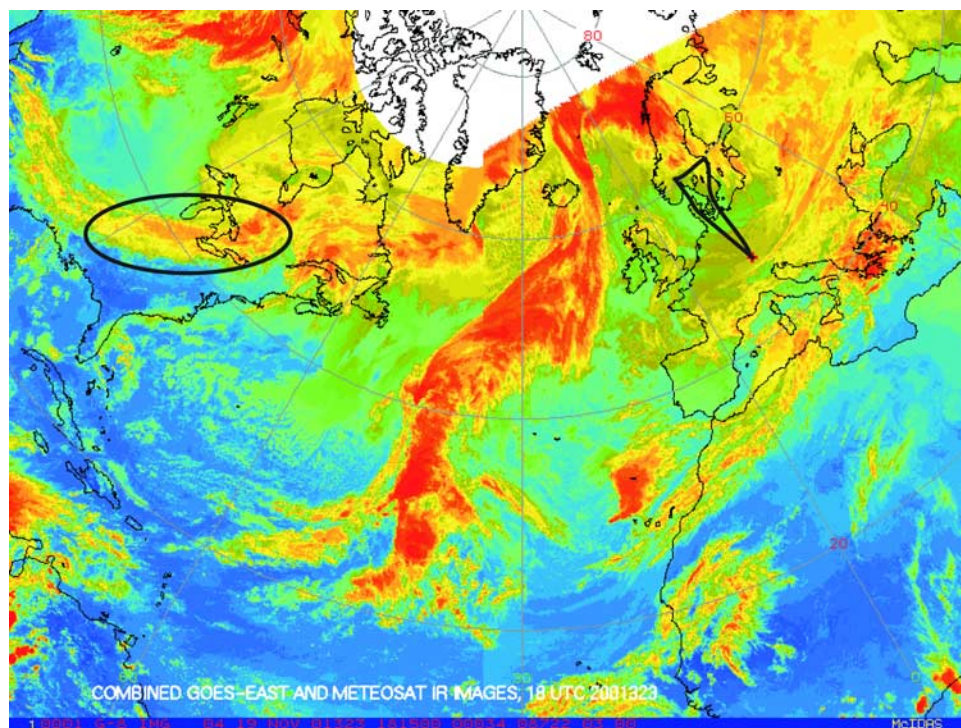


Figure 1. Combined GOES-EAST and METEOSAT infrared satellite image from 19 November 2001 (18 UTC). The flight track of the Falcon is superimposed (black triangle). The flight started/ended in Oberpfaffenhofen (Germany), and the aircraft made a stopover in Stockholm (Sweden). At the time of the airborne CONTRACE measurements over Scandinavia on 19 November 2001 the next cold front and associated warm conveyor belt pushed polluted air masses from the eastern United States out to the Atlantic (black ellipse).

refer to pressure altitude and all clock times refer to UTC if not given separately.

3.3. Model Description

[16] To simulate the transport of the North American pollution plumes, we used the Lagrangian particle dispersion model FLEXPART (version 4.4) [Stohl *et al.*, 1998] (<http://www.forst.tu-muenchen.de/EXT/LST/METEO/stohl/>). FLEXPART was validated with data from three large-scale tracer experiments in North America and Europe [Stohl *et al.*, 1998]. The model has been used in case studies of intercontinental air pollution transport [Stohl and Trickl, 1999; Forster *et al.*, 2004] and for studying its interannual variability over a 15-year period [Eckhardt *et al.*, 2003]. Details about the model simulations used in paper can be found in Stohl *et al.* [2003a, 2003b]. Therefore only a brief description is given here.

[17] For the tracer forecasts FLEXPART was driven with pressure-level data from the Aviation Model (AVN) of the National Center for Environmental Prediction (resolution 1° , 26 vertical layers, available every 3-hours). For the analyses FLEXPART was used with global data from the European Centre for Medium-Range Weather Forecasts (ECMWF) with a horizontal resolution of 1° , 60 vertical levels and a time resolution of 3 h. Wind fields with 0.5° resolution covering the domain 120°W to 30°E and 18°N to 66° were nested into the global data in order to achieve higher resolution over the region of main interest, i.e., North America, the North Atlantic and Europe. FLEXPART treats advection and turbulent diffusion by calculating the trajectories of a multitude of particles. Stochastic fluctuations, obtained by solving Langevin equations, are superimposed on the grid-scale winds to represent transport by turbulent eddies, which are not resolved in the ECMWF data. To

Table 1. Instrumentation of the Falcon Research Aircraft

Species	Technique	Detection Limit, nmol mol^{-1}	Sampling Time, s	Accuracy, %	Group	Reference
NO	chemiluminescence (CL)	0.005	1	10	DLR	Schlager <i>et al.</i> [1997], Feigl [1998], Ziereis <i>et al.</i> [1999]
NO _y	CL plus Au-converter	0.015	1	15	DLR	Feigl [1998]
O ₃	UV absorption	1	4	5	DLR	Schlager <i>et al.</i> [1997], Huntrieser <i>et al.</i> [1998]
CO	VUV fluorescence	2	1	3 nmol mol^{-1}	DLR	Gerbig <i>et al.</i> [1996, 1999]
SO ₂	Chemical Ionisation Mass Spectrometry (CIMS)	0.1	6.4	50	MPI-K	Arnold <i>et al.</i> [1997]
Acetone	Chemical Ionisation Mass Spectrometry (CIMS)	0.01	6.4	50	MPI-K	Wohlfrom <i>et al.</i> [1999]

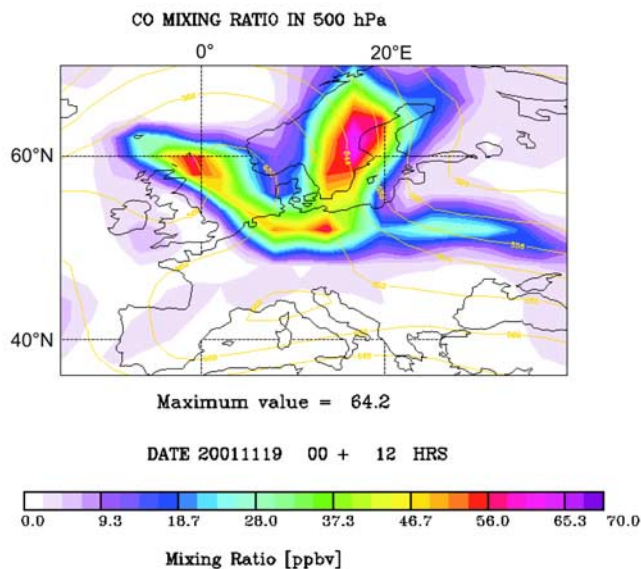


Figure 2. FLEXPART 12-hour forecast of a North American CO tracer at 5000 m a.s.l. for 19 November 2001 at 1200 UTC. Superimposed are isolines of the geopotential height at 500 hPa.

account for sub-gridscale convective transport, the convection scheme developed by *Emanuel and Zivkovic-Rothman* [1999] was applied.

[18] We calculated the transport of two passive tracers representing CO emissions from North America and Europe, taken from the EDGAR version 3.2 inventory [*Olivier and Berdowski*, 2001] for the year 1995. The simulation started on 28 October and ended on 28 November 2001. During this period, a total of 25 million particles were released between the surface and ~ 150 m above the ground at a constant rate, with the number of particles released in a particular grid cell being proportional to the emissions in that cell. In all

FLEXPART figures presented in this paper only the North American or European CO contribution (based on emission inventories) to total CO is shown.

4. The 19 November 2001 Case

4.1. Airborne Measurements With the Falcon Aircraft and FLEXPART Model Simulations

[19] The meteorological and chemical history of the North American pollution plume arriving over Scandinavia on 19 November has already to some extent been described by *Stohl et al.* [2003a]. For example the movement of the pollution plume over the North Atlantic is shown in Figures 3f–3j in *Stohl et al.* [2003a]. In this paper we focus on the impact of the North American pollution plume across Europe, and the flight from Germany to Scandinavia is described in detail next.

[20] The flight on 19 November began in Oberpfaffenhofen (southern Germany, 48°N , 11°E) at 10 UTC with destination, Stockholm, Sweden (Figure 1). The aircraft soon reached cruising altitude at 9.4 km. After about one hour the aircraft performed a stepwise descent over Denmark, and just south of Oslo, Norway, the lowest level was reached (1.7 km) at 12 UTC. Thereafter, the aircraft ascended stepwise to the east up to 6 km altitude and then landed in Stockholm. The stepwise pattern at lower levels over Scandinavia was based on the CO tracer forecasts, which indicated a pronounced North American CO plume as shown in Figure 2 for the FLEXPART model (see also MATCH-MPIC CO tracer forecast in Figure 13a in *Lawrence et al.* [2003]). The color bar in Figure 2 indicates only the North American CO contribution to total CO.

[21] The CO tracer taken from a subsequent simulation, using analysis rather than forecast meteorological data and interpolated to the flight track, confirmed that the North American plume was penetrated several times during the flight as shown in Figure 3 for the FLEXPART model (see

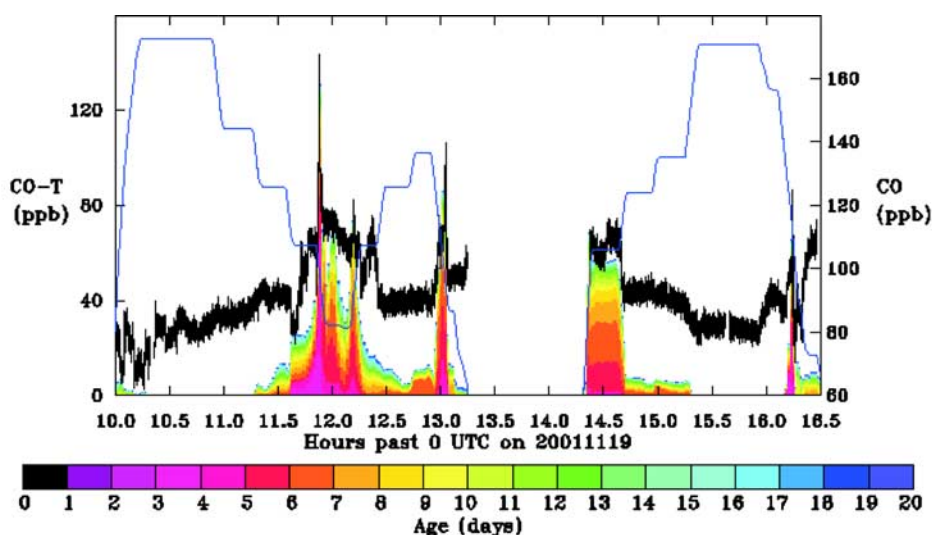


Figure 3. Age spectra of North American CO tracer obtained from backward model simulations with FLEXPART (colored bars) for the flight on 19 November 2001. Superimposed CO measured by the aircraft (black line) and aircraft altitude (blue line, relative units, maximum 9400 m) [from *Stohl et al.*, 2003a, Figure 8b].

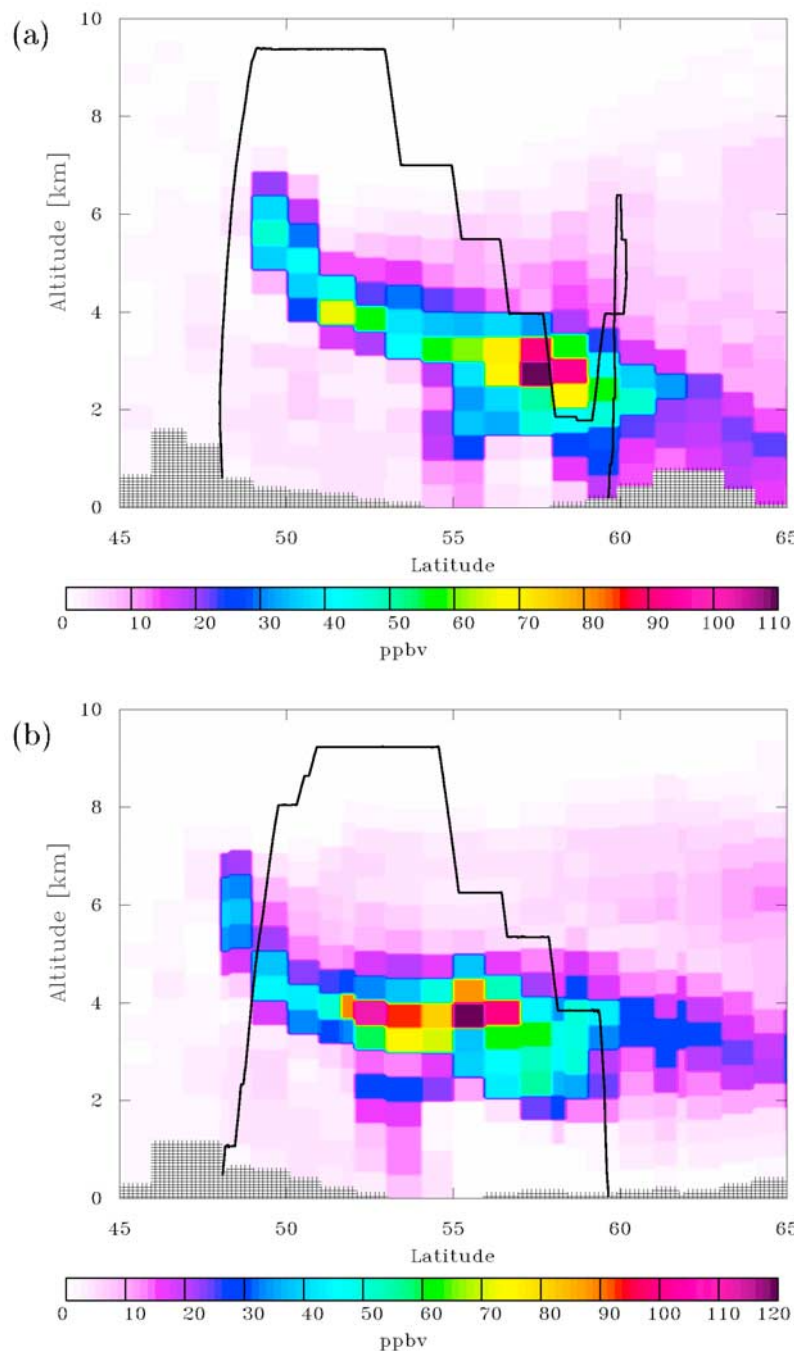


Figure 4. Vertical cross sections through the FLEXPART North American CO tracer field on 19 November 2001 (a) along 10°E at 1200–1300 UTC and (b) along a straight line between Stockholm and Oberpfaffenhofen (southern Germany) at 1400–1500 UTC. The aircraft altitude is superimposed (black lines) [from Stohl *et al.*, 2003a, Figure 5].

also MATCH-MPIC model results in Figure 15a in Lawrence *et al.* [2003]). In Figure 4a the vertical cross section through the FLEXPART North American CO tracer field along 10°E at 1200–1300 UTC is shown together with the altitude during the flight. The slanted structure of the pollution plume is clearly visible. The southernmost part of the plume, the leading edge, was located at 6 km altitude above southern Germany, while the northern part reached down to 2–4 km above central Scandinavia. The plume was penetrated 3 times during the flight (descent and ascent

close to Oslo, and descent close to Stockholm). The most pronounced CO tracer signal was found in the lower layer during the descent south of Oslo. Therefore this part of the flight was selected to present the airborne in situ measurements in more detail.

[22] In Figure 5a the vertical CO and O₃ profiles for the time of the descent near Oslo are shown (10 s mean values). In the mid and upper troposphere typical CO background values of $\sim 90 \text{ nmol mol}^{-1}$ were observed. In the lower troposphere (2–4 km) a pronounced layer with

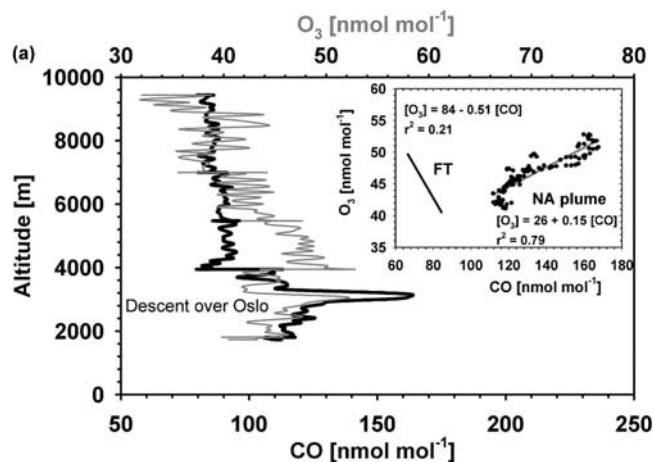


Figure 5a. Measured vertical profiles (10 s mean values) during the descent close to Oslo (Norway) on 19 November 2001 for CO (black line) and O₃ (gray line). Superimposed (upper right corner) are O₃-CO correlations in the free troposphere (FT, only regression line) and in the North American (NA) pollution plume.

CO up to almost 170 nmol mol⁻¹ was observed, which can be attributed to the North American pollution plume (Figures 3 and 4). CO was enhanced by ~100% in comparison to the typical background in 3 km (80–90 nmol mol⁻¹), which is a clearly observable signal even after 4 days of travel across the Atlantic. The vertical extension of the plume agrees very well with that of the FLEXPART CO tracer. The observed CO enhancement in the plume (~80 nmol mol⁻¹ above the background) was slightly lower than the value the FLEXPART model suggested (>110 nmol mol⁻¹).

[23] Further, the O₃ distribution was investigated for the descent south of Oslo. In general O₃ was negatively correlated with CO in the free troposphere (FT) above 4 km as shown in Figure 5a (upper right). For the free troposphere only the regression line is shown and not all single measurement points. In contrast, in the North American pollution plume (2–4 km) ozone increased from 43 to 53 nmol mol⁻¹ (i.e., by 23%). In this part of the plume H₂O was positively correlated with O₃ (not shown here) and increased from ~1.5 to 2.0 mmol mol⁻¹. The enhanced H₂O mixing ratio in the plume indicates that air has been uplifted from the more humid boundary layer and not transported downward from the drier upper troposphere - tropopause region. Furthermore, a positive O₃-CO correlation was observed in the plume (slope 0.15), which suggests that photochemical O₃ production must have been taken place. However, the season (late autumn), the northerly pathway of the polluted air mass passing Greenland and the transport in the lower-mid troposphere are factors which likely inhibit in-transit O₃ production. Ozone was probably already produced before the air mass was lifted over the Atlantic (see section 2). The likely source region was determined by the use of sophisticated backward modeling methods [Stohl *et al.*, 2003a]. The O₃ concentrations in the plume's source region over the eastern United States are investigated in section 5. Observations in the eastern United States confirmed that O₃ was strongly enhanced in the plume in comparison to the average. The meteorological conditions

were conducive for photochemical O₃ formation, which might explain the observed positive O₃-CO correlation in the North American plume over Scandinavia.

[24] Distributions of NO and NO_y (sum of all reactive, oxygen-containing nitrogen species: NO + NO₂ + PAN + HNO₃ + HNO₄ + NO₃ + N₂O₅ + aerosol nitrates) are also available for the descent south of Oslo (Figure 5b). As expected the short-lived trace gas NO is not enhanced in the North American pollution plume. The low NO values (below 0.05 nmol mol⁻¹) give no indications of freshly polluted air from nearby sources (Scandinavia) and further confirm the North American origin of the plume. In contrast, NO_y is elevated in the layer between 2 and 4 km with values of ~0.5–1 nmol mol⁻¹ in comparison to background values ~0.1–0.3 nmol mol⁻¹. The correlation between NO_y and CO in the free troposphere (FT) above 4 km and in the North American (NA) pollution plume (2–4 km) is presented in Figure 5b (upper right). For the free troposphere only the regression line is shown and not all single measurement points. Since both trace gases are emitted during combustion processes in the boundary layer, they are well correlated and generally decrease with altitude. The NO_y/CO slope depends on the age of a polluted air mass as suggested by Stohl *et al.* [2002b], who used data from NARE. NO_y can be removed by various processes whereas CO is approximately conserved on the timescales considered here. The NO_y/CO slope found in our North American pollution plume on 19 November was 0.008, which corresponds to an air mass age of 4–5 days since emission according to Figure 5 in Stohl *et al.* [2002b]. These findings agree well with the FLEXPART CO tracer simulations of the age of the pollution plume (Figure 3).

[25] Airborne SO₂ measurements are only available for the last part of the descent south of Oslo (just before and during the penetration of the pollution plume). Nevertheless, they further confirmed that the North American air mass after 4 days travel over the Atlantic was still polluted (Figure 5c). In the plume SO₂ mixing ratios up to 2.6 nmol mol⁻¹ were observed, which is distinctly higher

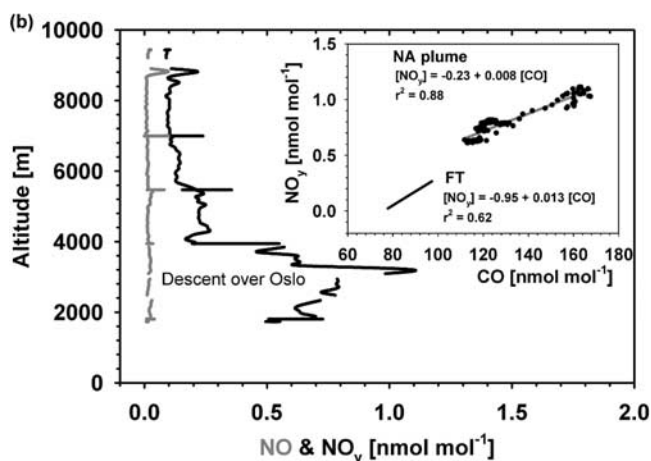


Figure 5b. Measured vertical profiles (10 s mean values) during the descent close to Oslo (Norway) on 19 November 2001 for NO_y (black line) and NO (gray line). Superimposed (upper right corner) are NO_y-CO correlations in the free troposphere (FT, only regression line) and in the North American (NA) pollution plume.

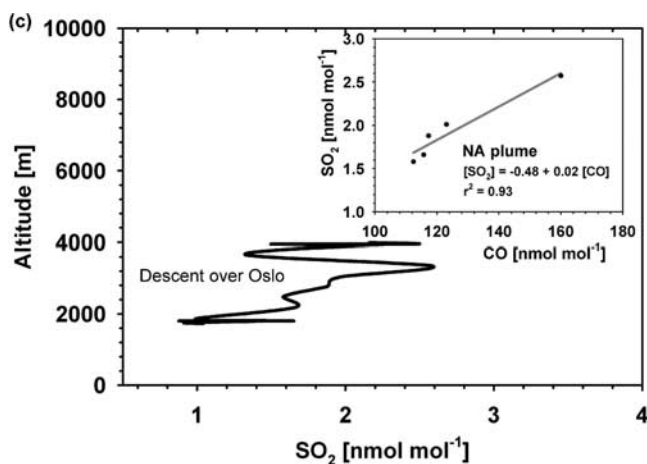


Figure 5c. Measured vertical profile (40 s mean values) during the descent close to Oslo (Norway) on 19 November 2001 for SO_2 (black line). Superimposed (upper right corner) is the SO_2 -CO correlation in the North American (NA) pollution plume.

than the typical background SO_2 of $\sim 0.5 \text{ nmol mol}^{-1}$. SO_2 shows a positive correlation with CO (Figure 5c); however, only a few measurement points are available due to the low SO_2 sampling rate. Acetone data are only available for the ascent over Oslo (same low sampling rate as for SO_2). Acetone increased up to 5 nmol mol^{-1} in the plume which is well above the background ($\sim 2 \text{ nmol mol}^{-1}$) and is also positively correlated with CO (Figure 5d).

[26] Both acetone and SO_2 in this plume are in the same range as previously reported by Arnold *et al.* [1997] for a North American pollution event. Arnold *et al.* observed enhanced SO_2 and acetone mixing ratios up to 3 nmol mol^{-1} during a flight in the north-eastern Atlantic in October 1993. These measurements indicate that North American sulphur emissions may contribute to the acid rain deposition over Europe as found by, e.g., Tarrason and Iversen [1992].

[27] The trace gas signatures observed during the subsequent ascent away from Oslo are not shown here in detail, since the structure was similar to the descent profile except that the plume was less pronounced ($\text{CO} < 120$, $\text{O}_3 < 52$, $\text{NO}_y < 0.6$, $\text{SO}_2 < 2.5$, and acetone $< 5.0 \text{ nmol mol}^{-1}$). The same is also true for the descent over Stockholm before landing ($\text{CO} < 140$, $\text{O}_3 < 52$, $\text{NO}_y < 0.7$, $\text{SO}_2 < 1.4$, and acetone $< 3.3 \text{ nmol mol}^{-1}$).

[28] On the return flight back to Germany the FLEXPART and MATCH-MPIC simulations show that the leading edge of the plume was penetrated before landing in Oberpfaffenhofen (between 4 and 6 km altitude) as presented in the Figures 3 and 4 for the FLEXPART model (see also Figure 15b in Lawrence *et al.* [2003]). Hours later and more than 1000 km to the south, the chemical signatures of the plume were similar to those found over Scandinavia ($\text{CO} < 120$, $\text{O}_3 < 52$, $\text{NO}_y < 0.7$, $\text{SO}_2 < 1.7$, and acetone $< 3.3 \text{ nmol mol}^{-1}$).

4.2. Dispersion of the North American Pollution Plume Over Europe

[29] From these first analyses we conclude that the North American pollution plume covered a large part of Europe on

19 November. In the following days the plume expanded from western Norway to western Finland, and from northern Scandinavia to the Alps as indicated in the FLEXPART simulations for the lowest model layer (from surface to 100 m agl) in Figures 6a–6f. According to the model simulations about half of Europe was covered by the North American pollution plume. However, the aircraft observations of the plume were made mainly between 2 and 6 km altitude and not at the ground where the pollutants may impact European air quality. We are therefore interested to know, whether this pollution plume from North America affected surface concentrations over Europe or not. The FLEXPART CO tracer distribution in the lowest model layer (Figures 6a–6f) suggests that a large part of Northern and Central Europe was affected in the time period 19 to 22 November. One CO tracer maximum passed over Norway (early morning 20 November), a second over the Alps (early morning 22 November), and a third over northern Finland (noon 21 November). The CO enhancements above the typical background (90 nmol mol^{-1}) in these maxima were $30\text{--}50 \text{ nmol mol}^{-1}$ which is well in agreement with the observations over Oslo $110\text{--}120 \text{ nmol mol}^{-1}$ CO in the lowest part of the NA pollution plume (Figure 5a). In section 6 we look more closely at the impact of the North American pollution plume on surface sites in the three mentioned regions. However, before we start to analyze trace gas signatures at European sites, we want to more thoroughly study the chemical conditions in the source region of the pollution plume over the eastern United States.

5. Chemical and Meteorological Measurements From the Eastern United States in November 2001

5.1. Horton: A CASTNET Sampling Station

[30] A rural site in the Appalachians (Horton, 920 m, 37.3°N , 80.6°W) was chosen to investigate the passage of pollution plumes over the eastern United States in November 2001. Horton, located on a mountain top, is one of many monitoring sites in the CASTNET (Clean Air Status and

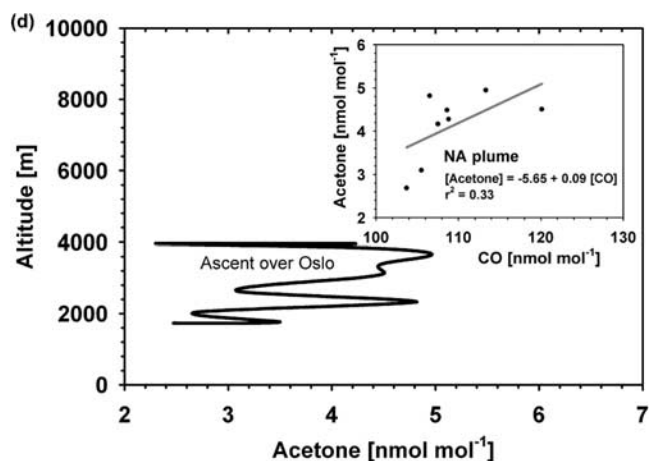


Figure 5d. Measured vertical profile (40 s mean values) during the ascent close to Oslo (Norway) on 19 November 2001 for acetone (black line). Superimposed (upper right corner) is the acetone-CO correlation in the North American (NA) pollution plume.

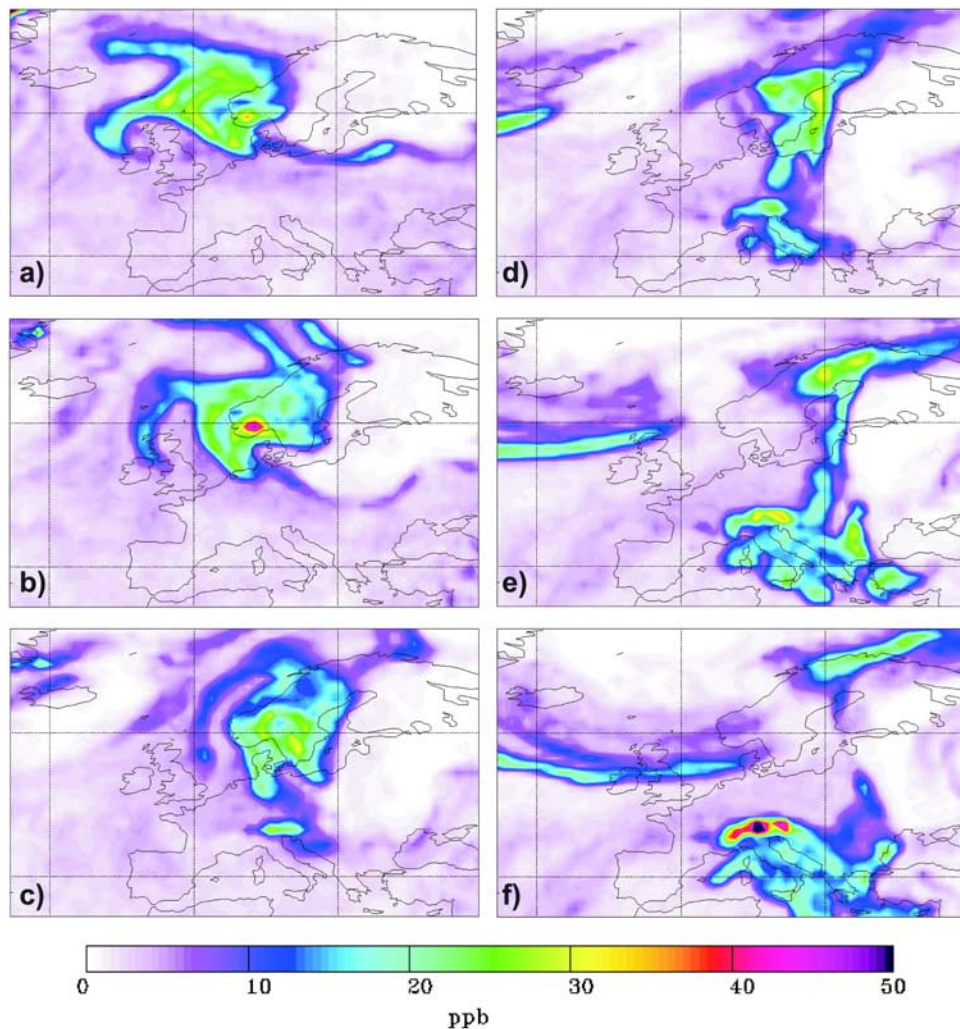


Figure 6. FLEXPART simulations of North American CO tracer for the lowest model layer (from surface to 100 m agl) showing the progress of the North American pollution plume over Europe (a) on 19 November 2001 at 12 UTC, (b) on 20 November at 00 UTC, (c) at 12 UTC, (d) on 21 November at 00 UTC, (e) 12 UTC, and (f) on 22 November at 00 UTC.

Trends Network) operated by the EPA (U.S. Environmental Protection Agency).

[31] Stagnant high-pressure conditions dominated the weather situation over the southern and eastern United States in October–November 2001. Precipitation data from the Horton site indicate a very dry period with no rain from 15 October to 24 November (not shown here). The low relative humidity combined with dry leaves on the ground created extreme fire danger in many eastern states. These autumn months are traditionally known as the wildfire season. The 2001 fire season was, however, not as extreme as in 2000, which was one of the worst seasons on record. More details about the November 2001 fires in the eastern United States can be found in the paper by *Stohl et al.* [2003a]. Simulations by *Stohl et al.* also showed that the enhancement in the CO tracer over the eastern United States could partly be explained by CO emissions from these fires. Due to the fires an extended haze layer built up over the eastern United States as remotely sensed, e.g., by the SeaWiFS instrument on 15 November (Figure 7).

[32] The SO₂ records from the Horton site show that the air was not only polluted by the smoke from the fires, but also by anthropogenic pollution, which accumulated to high levels during the stagnant weather conditions. Weekly samples of total SO₂ were analyzed for the time period July to December 2001 (not shown here). The period from 16 October to 20 November was notable, where total SO₂ was elevated over a whole month (10–20 μg m⁻³) in comparison to the mean total SO₂ of ~5 μg m⁻³ before and after this period.

[33] Furthermore, the high-pressure weather situation (strong insolation) and the burning fires (producing O₃ precursors) were favorable conditions for O₃ production. Especially in the southeastern parts of the United States, along the southern and western sides of the high-pressure systems, ozone was produced. Hourly O₃ records from Horton during November 2001 are presented in Figure 8. Ozone mixing ratios mainly vary between 20 and 60 nmol mol⁻¹. A number of longer lasting events with elevated O₃ (~40–60 nmol mol⁻¹) are recognizable: (1) 7–8 November, (2) 14–19 November

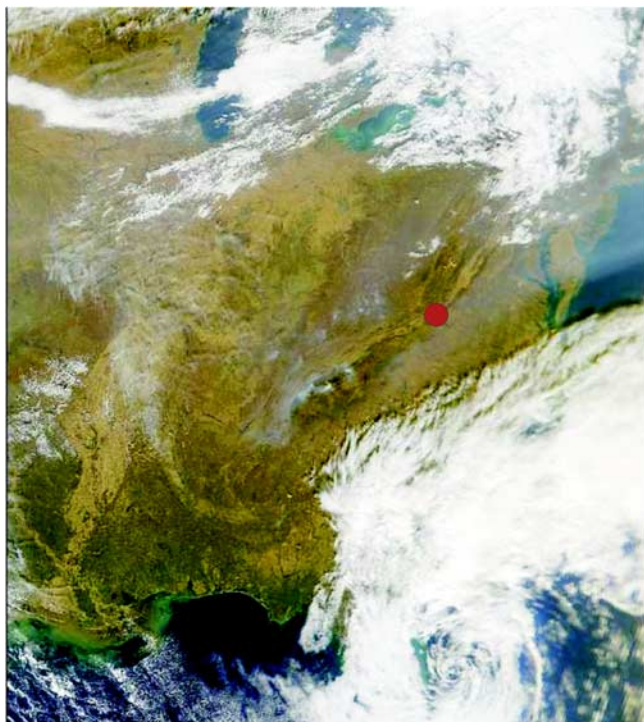


Figure 7. SeaWiFS satellite image from 15 November 2001. An extended polluted haze layer built up over the eastern United States since the end of October. Smoke from forest fires in Virginia, Kentucky, and West Virginia is visible, and the smoke even extended over the Chesapeake Bay. In the upper left corner an approaching cold front is visible. The red circle indicates the location of the Horton site.

(with interruptions), and (3) 23–24 November. In all these events the prevailing winds, ahead of cold fronts approaching from the west, pushed O_3 rich air masses from the southeastern parts of the United States to the Horton site. Traveling eastward, the polluted air masses were uplifted with warm conveyor belts and transported across the North Atlantic Ocean. During the CONTRACE period, four events that transported North American pollution layers over Europe were observed, which were all connected to these high O_3 episodes observed over the southeastern United States and at the Horton site ~ 3 – 5 days earlier. During most episodes of high O_3 , the FLEXPART-CO tracer was also enhanced at Horton as shown in Figure 8, which further confirms that O_3 was produced photochemically in highly polluted air masses with elevated CO.

[34] Finally, precipitation and O_3 data from Horton were analyzed for a period of three years, from 1999 to 2001. The dry period in autumn (October–November) occurs in all years. The seasonal O_3 cycle normally shows an O_3 maximum in late spring and early summer [Thompson, 1992] for the northeastern United States (40°N). However, at Horton a secondary O_3 maximum (~ 40 – 60 nmol mol^{-1}) occurred during the dry period in autumn for all three years. Before and after this period O_3 mixing ratios around 30 nmol mol^{-1} are measured. These analyses indicate that the O_3 conditions in autumn 2001 were not exceptional. This means that in autumn, when the storm track over the Atlantic is well developed, North American air masses

transported to Europe can be expected to contain elevated O_3 (and SO_2).

5.2. MOZAIC Flights

[35] To support the observations from the Horton site in the Appalachians, vertical O_3 profiles from several MOZAIC (Measurements of Ozone and Water Vapor by Airbus In-Service Aircraft) flights are presented here. Commercial flights with scientific instruments aboard have been carried out between North America and Europe for the MOZAIC program since 1994 [Marenco *et al.*, 1998]. Ozone profiles from four MOZAIC flights performed in November 2001 are selected. All times are in Eastern Standard Time (EST = UTC - 5 hours): (1) 14 November flight from Frankfurt to New York (descent ~ 12 EST, 40.6°N and 73.8°W), (2) 14 November flight from New York to Frankfurt (ascent ~ 16 EST, 40.6°N and 73.8°W), (3) 15 November flight from Washington to Vienna (ascent ~ 17 EST, 38.9°N and 77.5°W), and (4) 16 November flight from Boston to Frankfurt (ascent ~ 16 EST, 42.4°N , 71.0°W).

[36] Descent (1) around noon on 14 November in Figure 9a shows a typical vertical O_3 profile for the season with low mixing ratios at the ground (10 – 20 nmol mol^{-1}) and then a pronounced increase up to 2 km (40 – 50 nmol mol^{-1}). In the lower and mid troposphere O_3 continues to vary between 40 – 50 nmol mol^{-1} . In contrast, the ascents (2) and (3) in the late afternoon on 14 and 15 November (Figures 9b and 9c) indicate a pronounced layer with elevated O_3 up to 2 km altitude (peak mixing ratios up to almost 80 and 60 nmol mol^{-1} , respectively). In ascent (4) in the afternoon on 16 November (Figure 9d) the low level O_3 plume has disappeared as the polluted air mass has been swept out into the Atlantic by the cold front. Another O_3 plume is observed between 4 and 6 km. It is not very likely that this plume had any connection to the one previously mentioned. This elevated O_3 layer is observed behind the cold front and winds are prevailing from the northwest. No humidity data are available from the flight, but water vapor satellite images indicate dry air (probably downward transport from the stratosphere). Further, the

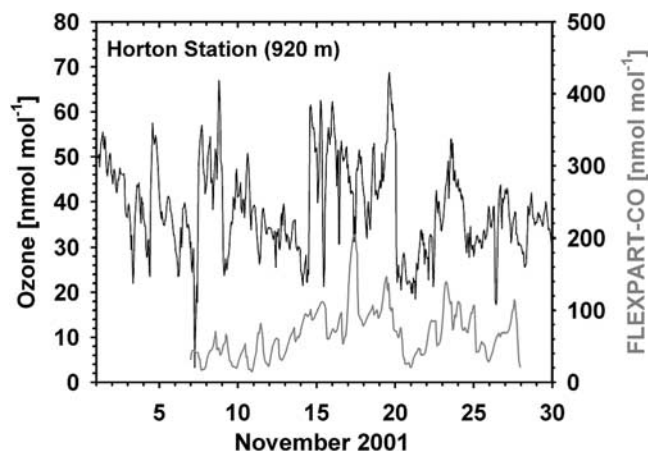


Figure 8. Hourly ozone records from the Appalachian mountain site Horton (920 m , 37.3°N , 80.6°W , in black), and hourly simulated values of the FLEXPART NA-CO tracer (in grey) for November 2001. Data source: EPA.

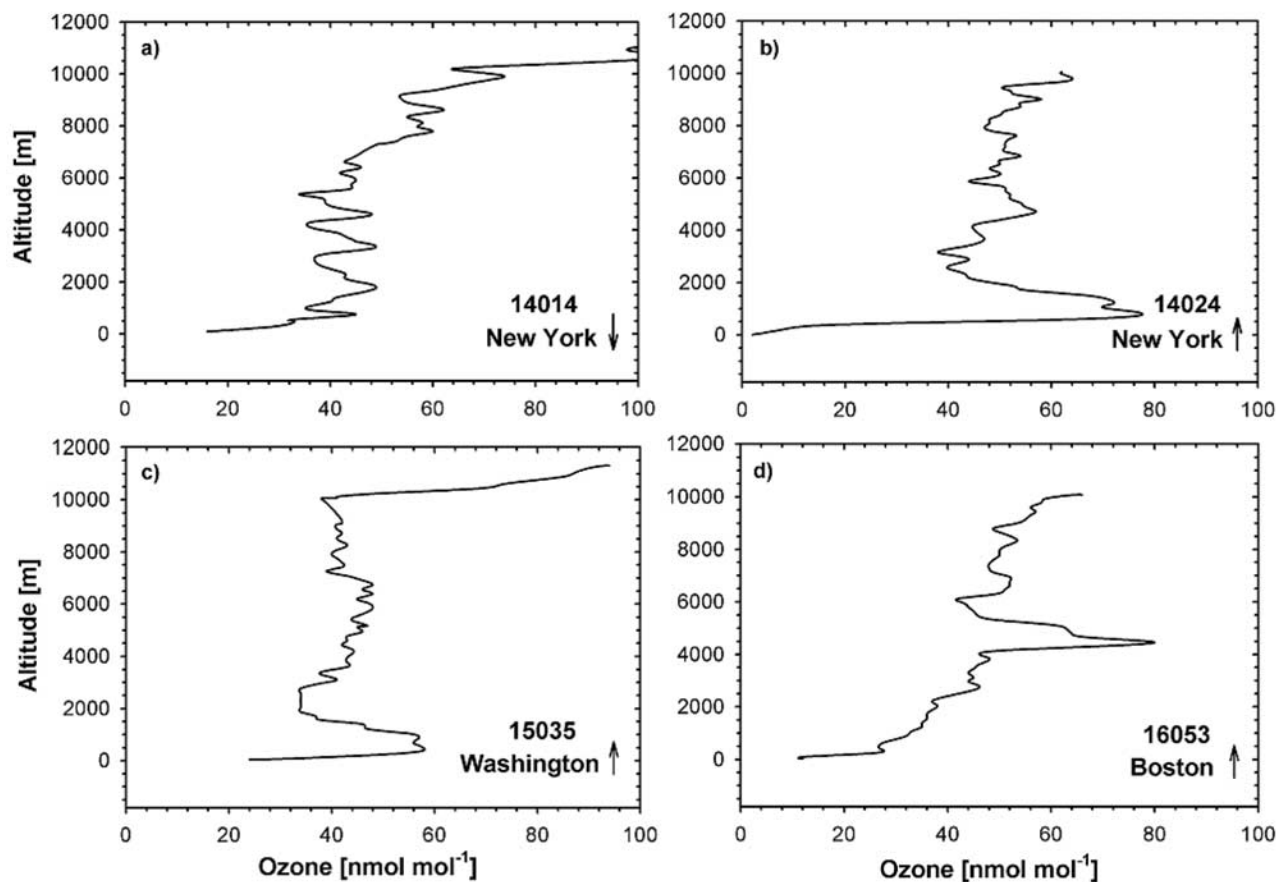


Figure 9. Ozone profiles from four MOZAIC flights performed in November 2001. All times are in Eastern Standard Time (EST = UTC - 5 hours): (a) 14 November flight from Frankfurt to New York (descent ~ 12 EST, 40.6°N and 73.8°W), (b) 14 November flight from New York to Frankfurt (ascent ~ 16 EST, 40.6°N and 73.8°W), (c) 15 November flight from Washington to Vienna (ascent ~ 17 EST, 38.9°N and 77.5°W), and (d) 16 November flight from Boston to Frankfurt (ascent ~ 16 EST, 42.4°N , 71.0°W).

tracer simulations with FLEXPART indicate that the main position of the pollution plume is already much further northeast at this time.

6. Chemical and Particle Measurements From European Surface Sites in November 2001

6.1. Ozone Measurements in Norway

[37] In addition to the airborne measurements, carried out with the Falcon aircraft over Oslo on 19 November, ground measurements from Norway were also analyzed since the FLEXPART simulations indicated that the North American pollution plume also affected the Norwegian surface (Figure 6b). Three EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe) ozone sites provided by NILU (Norwegian Institute for Air Research) were selected to study the passage of the North American pollution plume over Norway: (1) Voss (500 m, 60.6°N , 6.5°E) close to Bergen and the North Atlantic, (2) Hurdal (300 m, 60.4°N , 11.1°E) close to Oslo, and (3) Osen (440 m, 61.2°N , 11.8°E) close to Lillehammer and Sweden.

[38] In November 2001 the hourly O_3 records from these three Norwegian background sites mainly varied

between 10 and 40 nmol mol^{-1} (not shown here). The Voss site indicated a pronounced O_3 minimum down to $5\text{--}10 \text{ nmol mol}^{-1}$ for the time period when the NA plume passed over Oslo (19 November). However, in this case (below 500 m) the influence from the European pollutants dominated over the North American pollutants, and mainly caused the pronounced O_3 minima observed at the Norwegian sites. European emissions were accumulated in a stagnant high-pressure system over the British Isles and then advected to the Norwegian sites, in the time period 19 to 21 November, ahead of an approaching cold front from the west. A similar case, where European pollutants are advected at low levels ahead of an approaching cold front and North American pollutants are transported above, is described in section 7.

6.2. Ozone Soundings in Sodankylä (Finland)

[39] The North American CO plume separated into two branches over western Scandinavia as shown in the FLEXPART model simulations (Figures 6a–6f). One part of the plume moved southward toward the Alps and descended behind a trough into a surface anticyclone west of the trough, which is described in more detail in section 6.4 (Figure 6c). The other part of the plume moved to the

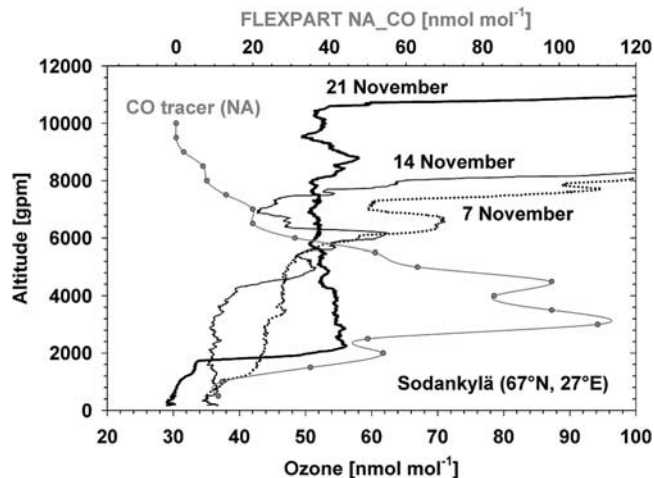


Figure 10. Three ozone soundings from Sodankylä (67.4°N, 26.6°E) in northern Finland from 7 (thin gray line), 14 (thin black line), and 21 (bold black line) November 2001. Data source: FMI-ARC. On 21 November an elevated ozone layer ($\sim 55 \text{ nmol mol}^{-1}$) is recognizable at 2–5 km which coincides with the maximum in the FLEXPART North American CO tracer (bold gray line with circles).

northeast and was uplifted by a new frontal system moving in from the Atlantic. Coincidentally, the plume passed the Finnish ozone sounding site Sodankylä (179 m, 67.4°N, 26.6°E) at noon on 21 November at the time of a weekly sounding (Figure 6e).

[40] In Figure 10, three ozone profiles from November 2001 are presented. On 21 November an enhancement in O_3 ($\sim 55 \text{ nmol mol}^{-1}$) is again recognizable at 2–5 km and below this level O_3 strongly decreases to 30 nmol mol^{-1} . Above this layer O_3 is almost constant (or even slightly decreasing) with height. The more typical O_3 increase with height, as shown in the other two profiles, is here missing in the troposphere. FLEXPART simulations of the North American CO tracer are also presented in Figure 10. The vertical CO tracer profile indicates a strong contribution from polluted North American air masses in the layer 2–6 km (up to $115 \text{ nmol mol}^{-1}$ CO), in accordance with the measured elevated O_3 layer. The ozone measurements from Sodankylä indicate that parts of the North American pollution plume investigated on 19 November over southern Norway could be identified a second time two days later over northern Finland.

6.3. Measurements With the Aerosol Lidar in Kühlungsborn (Germany)

[41] Under the framework of the EARLINET (A European Aerosol Research Lidar Network to Establish an Aerosol Climatology) project measurements with aerosol lidars were carried out intensively across Europe between February 2000 and February 2003. Fortunately, one of the observation days was 19 November 2001. Figures 4b and 6b show that the North American pollution plume passed over northern Germany in the evening on 19 November. The plume was observed with the aerosol lidar (Raman measurements at 532 nm) in Kühlungsborn (54.1°N,

11.8°E) in northern Germany. For further technical information on this lidar instrument the reader is referred to *Alpers et al.* [1999] and *Eixmann et al.* [2002].

[42] Figures 11a and 11b show vertical profiles of aerosol backscatter and extinction coefficient measured with the Kühlungsborn lidar on 19 November. The profiles are averaged over the time period 16:22–18:02 UTC. The vertical distribution of the extinction coefficient (Figure 11a) shows an enhancement in the lower troposphere between 2 and 4 km (mean value 0.02 km^{-1}), which can be attributed to aerosols in the North American pollution plume. The vertical CO tracer profile from FLEXPART simulations (Figure 11a, right) indicates a strong contribution from polluted North American air masses in the layer 2–4 km (up to $105 \text{ nmol mol}^{-1}$ CO). In addition, the vertical profile of the backscatter coefficient (Figure 11b) shows enhanced values from the ground up to 4.5 km. In the layer between 2 and 4 km values around $0.00015 \text{ km}^{-1} \text{ sr}^{-1}$ are measured, which is about the same range as reported by *Eixmann et al.* [2002] for another case with continental aerosols from North America observed over Germany. These measurements indicate that an aerosol layer can persist in the free troposphere for more than 4–5 days in accordance with the findings of *Eixmann et al.* [2002].

6.4. Chemical Measurements in the Alpine Region

[43] One branch of the North American pollution plume moved toward the Alps and descended behind a trough. The pollution was carried down to the surface by large-scale subsidence and mountain waves over the Alps. The 6-hourly FLEXPART tracer distributions for the lowest model layer (surface to 100 m agl) indicate that the North American pollution plume approached the Alpine region during the morning of 20 November 2001 (Figure 6c). A maximum in the North American CO tracer is present over eastern Switzerland, western Austria and northeastern Italy at 12 UTC. Over the next few days the North American CO tracer in that region slightly decreased and then increased again. The highest CO tracer concentrations in the Alpine region ($>50 \text{ nmol mol}^{-1}$ above background) occurred between 00 and 06 UTC on 22 November (Figure 6f). At this time the next North American pollution plume (case 22 November) approached northwestern Europe as an elongated streamer stretching from Ireland to Denmark. However, this plume traveled rapidly over Europe in a west-east direction and did not affect ground levels in the Alpine region (see also section 7).

[44] Four elevated rural sites, partly in the free troposphere, were selected to study the passage of the North American pollution plume in the Alpine region: in Germany (1) the Zugspitze mountain top site (2962 m, 47.4°N, 11.0°E) and (2) Hohenpeißenberg (976 m, 47.8°N, 11.0°E), and in Switzerland (3) the Jungfrauoch mountain top site (3580 m, 46.6°N, 8.0°E), and (4) Arosa (1840 m, 46.8°N, 9.7°E).

6.4.1. Zugspitze Mountain Site

[45] A combination of chemical measurements from Zugspitze and FLEXPART tracer simulations of North American and European CO was used to analyze the passage of the North American pollution plume. The November 2001 time series of CO, O_3 , NO_x , NO_y , and

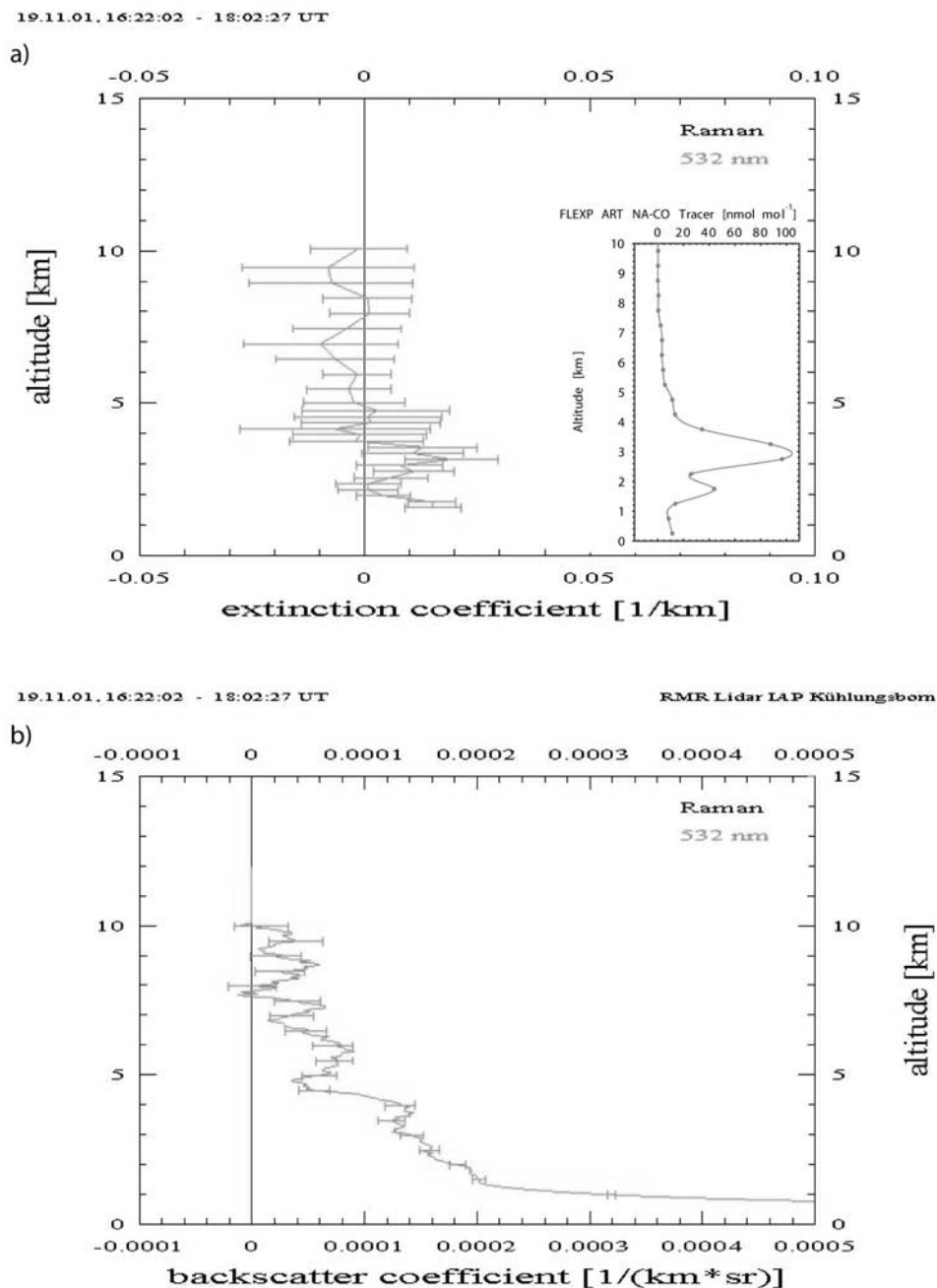


Figure 11. Vertical profiles of (a) the extinction coefficient and (b) the aerosol backscatter measured with the Kühlungsborn lidar (54.1°N, 11.8°E) in northern Germany on 19 November. The profiles are averaged over the time period 16:22–18:02 UTC. An elevated aerosol layer is recognizable at 2–4 km which coincides with the maximum in the FLEXPART North American CO tracer (bold gray line with circles).

⁷Be (Beryllium-7) from Zugspitze are shown in Figure 12a. ⁷Be is a good tracer to mark the time periods of stratospheric air masses affecting the Zugspitze site. The average ⁷Be for November (1996–1998) is 4.6 mBq m⁻³ according to Gerasopoulos *et al.* [2001], which is close to the November 2001 value (4.5 mBq m⁻³). Reiter *et al.* [1984] used a 30% increase in ⁷Be values against the monthly mean as a criterion for air masses of stratospheric origin. For Zugspitze this means that if ⁷Be exceeds 6 mBq m⁻³ stratospheric air masses may be present. However,

during the passage of the North American pollution plume ⁷Be mainly varied between 4.9 and 5.1 mBq m⁻³, which indicates no pronounced stratospheric influence. Stohl *et al.* [2000] used an even higher threshold of 8 mBq m⁻³ to identify air masses of stratospheric origin in the Alpine region.

[46] The passage of the North American pollution plume can be most clearly seen in the CO records from Zugspitze (red line in Figure 12a). A pronounced increase in CO is observable during two days from 20 November 06 UTC to

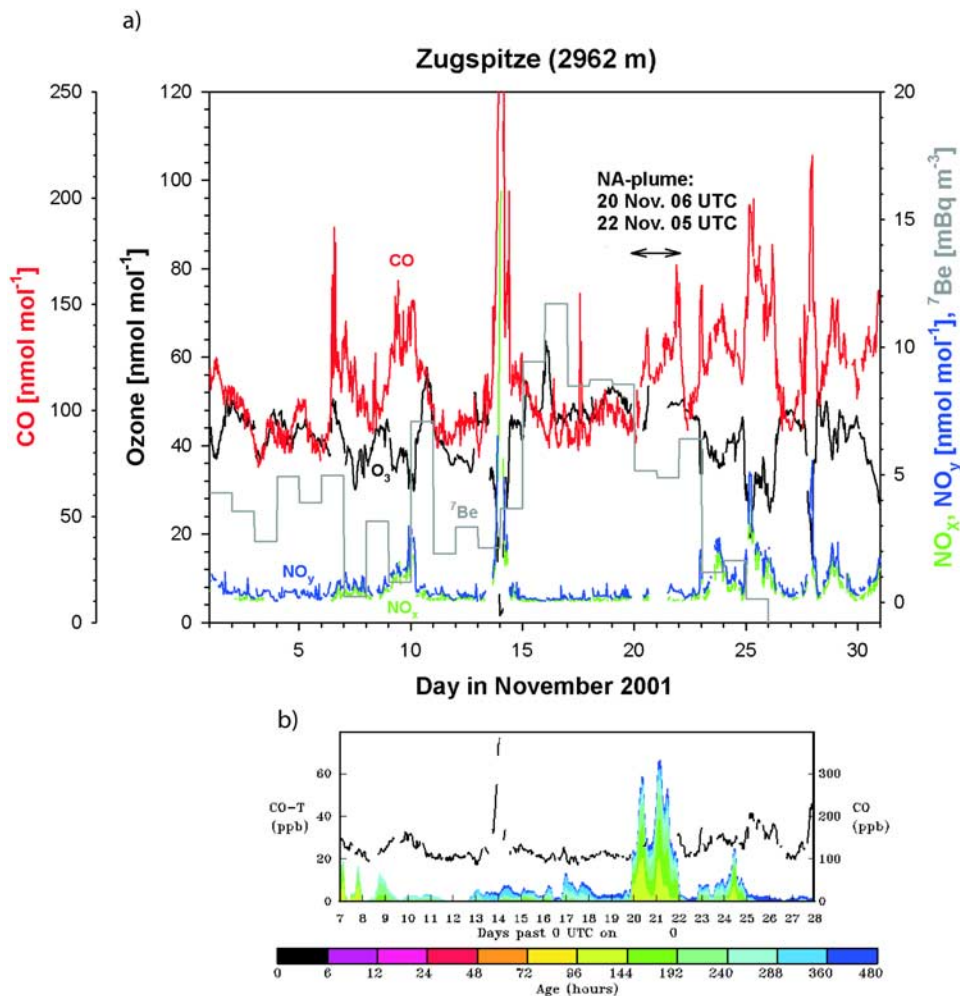


Figure 12. (a) Time series of measured CO (red), O₃ (black), NO_x (green), NO_y (blue), and ⁷Be (gray) from the Zugspitze site (2962 m, 47.4°N, 11.0°E) from November 2001. Below (b) the age spectra of the North American CO tracer (axis left) obtained from backward model simulations with FLEXPART (colored bars) for November 2001 for Zugspitze is shown together with measured CO (black line, axis right).

22 November 05 UTC (average CO is 126 nmol mol⁻¹). Just before and after the event CO varied between 90 and 100 nmol mol⁻¹. During the whole pollution event three CO peaks are visible. The first two almost reach 140 nmol mol⁻¹, and the third and strongest peak reaches 168 nmol mol⁻¹ (values in accordance with the airborne measurements presented in section 4). In comparison, the average CO mixing ratio at Zugspitze in November 2001 is 114 nmol mol⁻¹. In Figure 12b the FLEXPART tracer simulations for Zugspitze from 7 to 28 November 2001 are shown. The contribution of the North American CO tracer (lowest panel in color) is highest in the time period 20 to 22 November in accordance with the CO measurements. The three-peak structure is also visible in the simulations, however the second peak is strongest. The simulated CO enhancement above the typical background (~90 nmol mol⁻¹) at the time of these three peaks varies between 50 and 70 nmol mol⁻¹, which is in good agreement with the observations mentioned above. Furthermore, during the passage of the aged North American

pollution plume NO_y and NO_x mixing ratios were low (<0.7 nmol mol⁻¹ and <0.3 nmol mol⁻¹, respectively).

[47] Unfortunately, due to technical problems with the O₃ instrument at the Zugspitze site, there are gaps in the O₃ records during the passage of the North American pollution plume. However, at the beginning of the event measurements are available which show a pronounced positive correlation between O₃ and CO (in accordance with the airborne measurements). In the first CO peak, ozone was about 54 nmol mol⁻¹, which is in the range of the highest O₃ records of the month at Zugspitze, if the stratospheric events on 10 and 15–16 November (according to the ⁷Be records) are excluded. At the Zugspitze site the monthly mean O₃ mixing ratio of November 2001 was 42 nmol mol⁻¹. This value is in good agreement with long-term records of O₃ (since 1978) for November (41.6 nmol mol⁻¹) as published by Carnuth *et al.* [2002, Table 2]. During the pollution event the mean O₃ mixing ratio was 50 nmol mol⁻¹, corresponding to an O₃ increase of 8 nmol mol⁻¹ (19%). In Table 2 a summary of the plume's CO and O₃ mixing ratios and mean

Table 2. CO and O₃ Records From Alpine Sites for November 2001

Site	Peak CO in NA Plume, nmol mol ⁻¹	Mean CO in NA Plume, nmol mol ⁻¹	Mean CO in Nov. 2001, nmol mol ⁻¹	Peak O ₃ in NA Plume, nmol mol ⁻¹	Mean O ₃ in NA Plume, nmol mol ⁻¹	Mean O ₃ in Nov. 2001, nmol mol ⁻¹	ΔO ₃ (Plume-Mean), nmol mol ⁻¹	ΔO ₃ (Plume-Mean), %
Zugspitze (2962 m)	168	126	114	54	50	42	+8	+19
Jungfraujoch (3580 m)	127	116	124	51	49	43	+6	+14
Arosa (1840 m)	158	146	166	49	48	36	+12	+33

values for November 2001 is presented for Zugspitze and for comparison measurements from Jungfraujoch and Arosa were added. Mean CO varied between 116 and 146 nmol mol⁻¹ in these plumes which is in good agreement with the FLEXPART simulations in Figure 6f (30–50 nmol mol⁻¹ added to the typical background of 90 nmol mol⁻¹). The trace gas signatures from Hohenpeißenberg, Jungfraujoch, and Arosa are described in more detail next.

6.4.2. Hohenpeißenberg Site

[48] Hohenpeißenberg is located about 40 km north of Zugspitze. During nighttime this site is frequently situated above the boundary layer in the free troposphere. During daytime, when the top of the boundary layer rises, Hohenpeißenberg frequently gets exposed to local pollution. We selected this site to investigate whether the North American pollution plume could impact trace gas signatures in the boundary layer. In Figure 13a, trace gas records at Hohenpeißenberg from 20 to 22 November 2001 are shown. This time period was selected in accordance with the passage of the pollution plume at Zugspitze.

[49] The FLEXPART tracer simulations for Hohenpeißenberg in Figure 13b show very low North American CO mixing ratios (range 3–11 nmol mol⁻¹), and no pronounced agreement with the variations in the CO measurements (Figure 13a). The strongest influence of North American CO is simulated for the morning hours on 22 November. However, at this time the simulations also indicated a much stronger influence from European CO (not shown). The passage of a fresh European CO pollution plume was observed in the measurements, which contained elevated CO (270 nmol mol⁻¹) and NO_y (19 nmol mol⁻¹), and low O₃ (5–10 nmol mol⁻¹) mixing ratios due to titration by NO. This case is further discussed in section 7.

[50] Overall, the FLEXPART tracer simulations indicate almost no impact of the North American plume on the Hohenpeißenberg site. The major part of the plume probably passed above the site. However, let us here assume that the part of the North American plume, that passed the Zugspitze site, also penetrated down to Hohenpeißenberg and mixed with boundary layer air. The mean CO, O₃, and NO_y mixing ratios in the North American plume that passed Zugspitze (in the time period 20 November 06 UTC to 22 November 05 UTC) were 126, 49, and 0.4 nmol mol⁻¹, respectively. The impact of the North American pollution plume at Hohenpeißenberg would be very different from the impact on the free tropospheric site Zugspitze. From the mean CO, O₃, and NO_y mixing ratios for November 2001 at Hohenpeißenberg (157, 26, and 6.8 nmol mol⁻¹, respectively), we would expect a pronounced decrease in CO and NO_y, and an increase in O₃ during the passage of plume. However, due to the mixing with local polluted air masses (high NO), the excess in O₃ would probably soon decrease down to

ambient mixing ratios due to titration by NO. Overall, the aged North American pollution plume would be “cleaner” with respect to CO and NO_y than the average local pollution situation at the Hohenpeißenberg site, and excess O₃ would probably not persist over a long time like the observations in the free troposphere indicate.

[51] Furthermore, the seasonal impact of U.S. emissions on the Hohenpeißenberg site has recently been investigated in CTM simulations carried out by *Wild and Akimoto* [2001]. The enhancement in three-hourly O₃ mixing ratios was most pronounced between February and May. In summer, when O₃ pollution is of greatest concern, the impact from U.S. emissions was low. A secondary maximum in three-hourly O₃ mixing ratios was found in autumn (October–November). The repeated high O₃ episodes over the eastern United States in autumn as observed during CONTRACE and the strengthening storm track over the Atlantic in this season may explain the secondary maximum in three-hourly O₃ mixing ratios simulated for Hohenpeißenberg.

6.4.3. Jungfraujoch Mountain Site

[52] The passage of the North American pollution plume is also seen in the trace gas records from the mountain top site Jungfraujoch in the time period 20 and 22 November (not shown here). The chemical signatures agree well with the observations from Zugspitze and the three-peak CO structure is also present, however, the peak values are lower (120–130 nmol mol⁻¹) and also the mean CO mixing ratio in the plume (116 nmol mol⁻¹). NO and NO_x mixing ratios are low (<0.1 nmol mol⁻¹), as expected in an aged polluted air mass. Ozone is rather constant during this event with mixing ratios ~48–50 nmol mol⁻¹. In comparison, the mean O₃ for November 2001 at this site was 43 nmol mol⁻¹ (Table 2). At the Jungfraujoch site the North American pollution event also ranks among the highest O₃ events of the month. It lies between the 75th and 90th percentiles of the daily O₃ maxima in November at Jungfraujoch, taken from a long-term study by *Weber and Prévôt* [2002].

6.4.4. Arosa Site

[53] Arosa is also well-known as a “European representative” background O₃ site [*Pochanart et al.*, 2001]. This study, however, shows that long-range transport of pollution from North America can enhance the typical background O₃ at this site significantly. Recent investigations of the ozone seasonal cycle at Arosa by *Pochanart et al.* [2001] also suggest that the background O₃ at this site is influenced by North American pollution.

[54] The North American plume reached the Arosa site later (evening on 20 November) and lasted longer (late morning on 22 November) in comparison to the observations at the mountain sites Zugspitze and Jungfraujoch, and partly in comparison to the FLEXPART NA-CO tracer

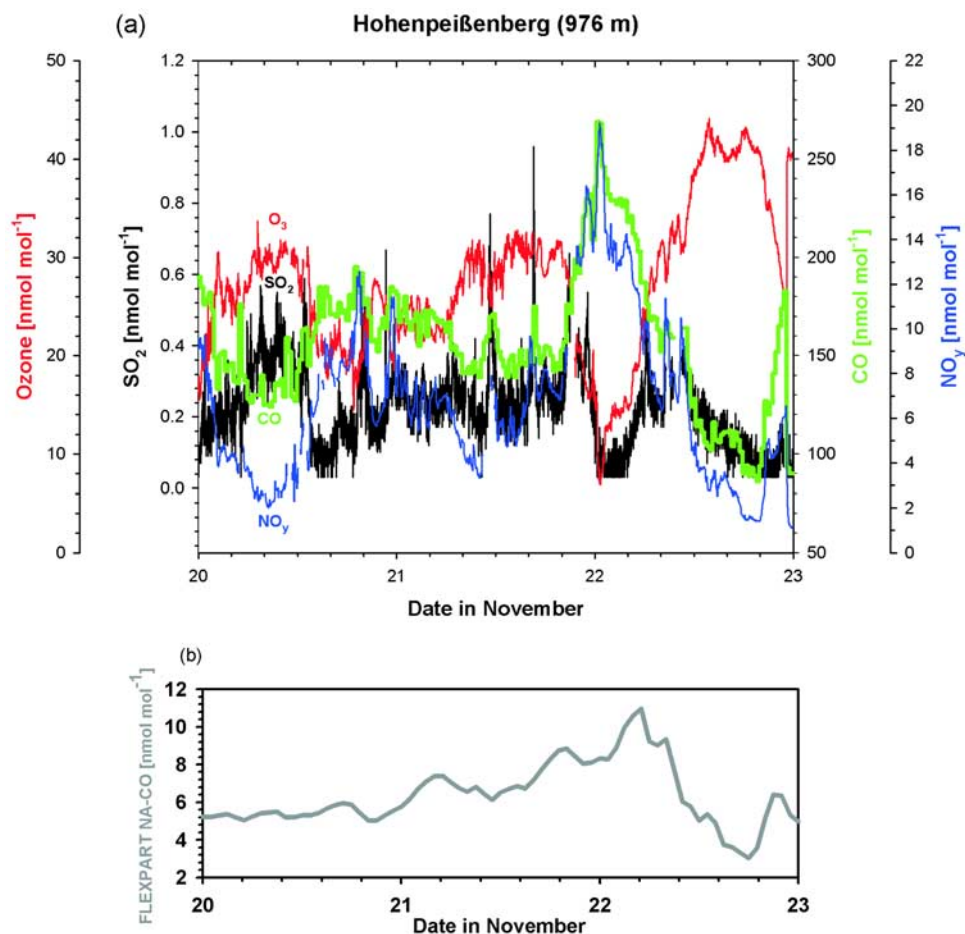


Figure 13. (a) Time series of measured CO (green), NO_y (blue), O₃ (red), and SO₂ (black) from the Hohenpeißenberg site (976 m, 47.8°N, 11.0°E) from 20 to 22 November 2001. The time period was selected in accordance with the passage of the North American pollution plume at Zugspitze. Below (b) FLEXPART North American CO tracer for November 2001 for Hohenpeißenberg.

simulations for Arosa (Figure 14). During the passage of the plume CO records from Arosa are rather constant $\sim 150 \text{ nmol mol}^{-1}$, which is slightly lower than the CO average at this site (Table 2). In contrast, the highest O₃ mixing ratios of November 2001 (49 nmol mol^{-1}) occurred during this event (Figure 14). During the two days of the plume's passage mean O₃ was $48 \pm 1 \text{ nmol mol}^{-1}$, which is distinctly higher than the mean for November 2001 (36 nmol mol^{-1}) and the mean for November from 1989–1991/1996–1997 (38 nmol mol^{-1}) as reported by Pochanart *et al.* [2001]. Ozone was on average elevated by 12 nmol mol^{-1} (33%) during this event, which is the largest O₃ enhancement for any Alpine site investigated in this study (Table 2). Ozone is normally lower at this site than at Zugspitze and Jungfrauoch, due to the general O₃ increase with increasing altitude. The ozone variability at Arosa was also observed in the O₃ records from Davos (1640 m, 46.8°N, 9.8°E), located close to Arosa but 200 m lower (not shown here).

[55] A number VOCs were also measured in November 2001 at the Arosa site. The ratio of toluene to benzene can be used to estimate the age of polluted air masses [Rappenglück and Fabian, 1999]. The lifetimes of benzene

and toluene with respect to average OH mixing ratios (τ_{OH}) are 2.4 and 0.5 days, respectively. At the beginning of the pollution event (early morning on 21 November) the lowest toluene/benzene ratio (0.32) of the whole month was estimated (benzene $49.5 \text{ pmol mol}^{-1}$ and toluene $15.7 \text{ pmol mol}^{-1}$). During daytime on 21 November the ratio increased to more common values (~ 1) due to the mixing with more freshly polluted air (mean toluene/benzene ratio for November 2001 was 1.1). The highest benzene and toluene mixing ratios measured were 65.6 and $75.9 \text{ pmol mol}^{-1}$, respectively. At the end of the pollution event (early morning on 22 November) again a very low toluene/benzene ratio (0.44) was estimated (benzene $55.5 \text{ pmol mol}^{-1}$ and toluene $24.3 \text{ pmol mol}^{-1}$). These low toluene/benzene ratios observed in the nighttime during the passage of the North American plume further confirm that the polluted air mass was aged.

7. The 22 November 2001 Case: Second North American Pollution Event Over Europe

[56] In the last section 6.4 the dispersion of a North American pollution plume over Europe (case 19 November)

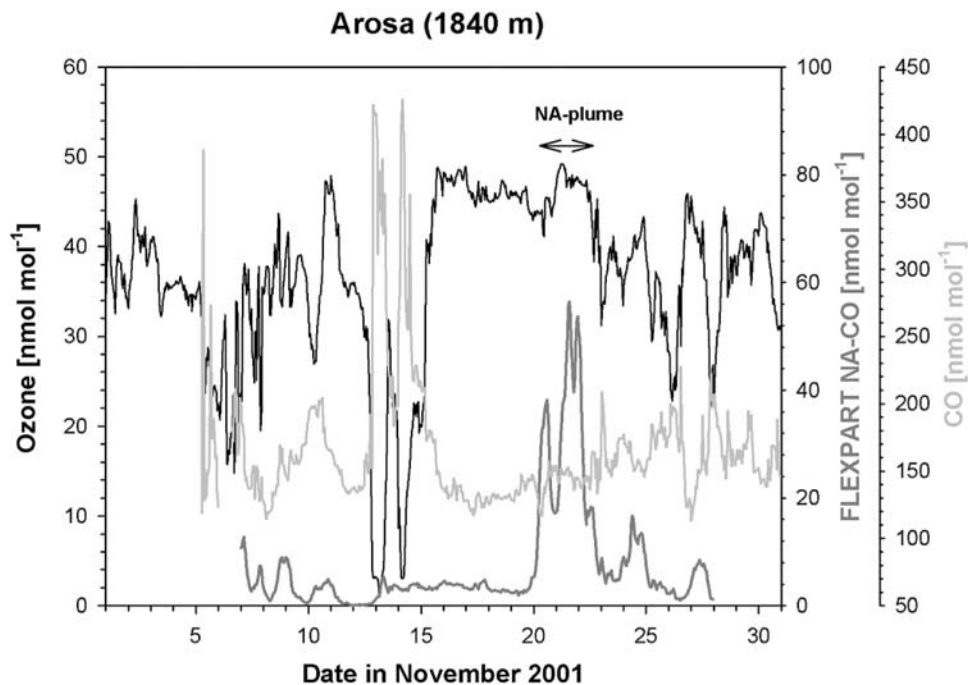


Figure 14. Ozone (black) and CO (light gray) records from the Arosa site (1840 m, 46.8°N, 9.7°E). Superimposed are hourly simulated values of the FLEXPART NA-CO tracer (dark gray) for November 2001.

was described in large detail. It was mentioned that a second North American pollution plume (case 22 November) approached northwestern Europe as an elongated streamer stretching from Ireland to Denmark (Figure 6f). In this section we briefly describe this second North American pollution event over Europe (case 22 November) which has a more complicated behavior since it was layered above fresh European (EU) emissions. The different chemical characteristics of the NA and EU air masses are briefly described here. Without the use of FLEXPART CO tracer forecasts and the knowledge of the tracer correlations in the North American pollution plume (case 19 November) the contribution from North American emissions to this second pollution plume over Europe had probably been overseen.

[57] At the time of the airborne CONTRACE measurements over Scandinavia on 19 November 2001 the next cold front and associated warm conveyor belt pushed polluted air masses from the eastern United States out to the Atlantic (Figure 1). This cold front was more pronounced and stronger than the one passing on 15 November. The O_3 measurements at the Horton site in the Appalachian mountains show a strong decrease in O_3 after the frontal passage (Figure 8). According to the FLEXPART CO tracer forecasts used for the flight planning, this second North American pollution plume (Figure 15a) reached Europe on 22 November and moved rapidly from the west (British Isles) to the east (Poland). The plume passed over Oberpfaffenhofen (southern Germany) in the late afternoon and was penetrated by the aircraft at around 16 UTC. The chemical situation was more complicated than in the previous case study. A strong cold front passed over Central Europe at the time of the passage of the North American plume. The FLEXPART CO tracer forecasts used for the flight planning indicated that this

front lifted European pollution to levels of about 3 km (Figure 15b). The North American pollution was located above the European pollution layer at around 6 km (Figure 16). The airborne measurements with the Falcon aircraft confirmed these forecasts. During the descent to Oberpfaffenhofen elevated CO was measured between 1 and 6 km altitude (Figure 17). Two major CO plumes are recognizable. The lower plume (2–5 km) could be attributed to the fresh European emissions and the upper one (5–6 km) to North American emissions according to the FLEXPART forecasts (Figure 16). CO mixing ratios are similar in both plumes (up to 160 nmol mol⁻¹), however, the O_3 -CO correlations show a distinct difference between the two air masses (Figure 17). In the North American air mass O_3 and CO are slightly positively correlated (slope 0.08) and O_3 increased by about 10 nmol mol⁻¹ from 40 to 50 nmol mol⁻¹. In contrast, in the European air mass O_3 and CO are negatively correlated (slope -0.50). Ozone decreased by about 20 nmol mol⁻¹ from 40 to 20 nmol mol⁻¹. Ozone is low in the European air mass due to titration by NO [Parrish *et al.*, 1998]. The NO_y -CO correlation also showed distinct differences between the two air masses (not shown here). The NO_y /CO slope was 0.010 in the North American air mass, which suggests an air mass age of 4 days since emissions according to Stohl *et al.* [2002b]. In the more freshly polluted European air mass the NO_y /CO slope was much steeper (0.135).

8. Discussion

[58] In this paper we investigated two North American pollution plumes that were transported over the North Atlantic to Europe in the free troposphere. Furthermore,

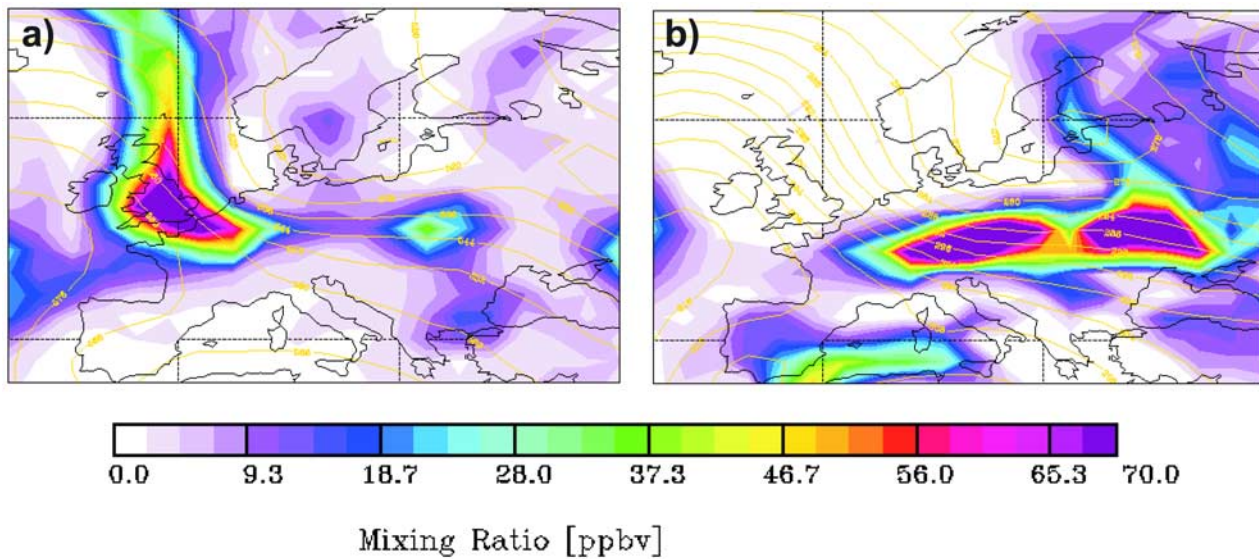


Figure 15. FLEXPART 15-hour forecasts of (a) a North American CO tracer at 5000 m a.s.l. and (b) a European CO tracer at 3000 m a.s.l. for 22 November 2001 at 1500 UTC. Superimposed are isolines of the geopotential height at 500 and 700 hPa, respectively. A second North American pollution plume (Figure 15a) reached Europe and moved rapidly from the west (British Isles) to the east (Poland). European pollution (Figure 15b) was uplifted over Germany.

North American pollution plumes can also be exported to Europe in the boundary layer as mentioned in the introduction. However, what is the impact of these plumes on European ozone distribution?

[59] Parrish *et al.* [1998, 2000] observed that in winter anthropogenic pollution reduces O_3 in the continental outflow from North America if it is advected into the marine boundary layer. The photochemical O_3 loss is here driven by sunlight, OH in the presence of elevated water vapor, and low NO mixing ratios.

[60] Jaffe *et al.* [2003] investigated a number of episodes of transatlantic air pollution transport and found a large variability in the O_3 distribution from case to case. Ozone was only enhanced in the pollution plumes (sometimes exceeding even 80 nmol mol^{-1}) if the transport took place in the free troposphere and in absence of mineral dust. Transport in the boundary layer prevented the import of elevated ozone. Jaffe *et al.* suggested that something in the marine boundary layer destroys O_3 in transit. Both OH and halogen radicals are candidates which may react and destroy ozone [Dickerson *et al.*, 1999]. Recent studies over the eastern North Atlantic in spring by Reeves *et al.* [2002] further suggest that photochemical O_3 loss is dominating in the marine boundary layer ($\sim 0.5 \text{ nmol mol}^{-1}$ per hour).

[61] If we apply the findings by Jaffe *et al.* [2003] to the transatlantic air pollution transport this means that only if a North American pollution plume is uplifted and transported in the free troposphere over the Atlantic, ozone may be enhanced in the plume (depending on the O_3 situation in the source region). This enhanced O_3 only affects ground sites in Europe if it descends into the boundary layer (seldom the case) or passes the Alpine region where the mountain sites reach into the free troposphere. Stohl *et al.* [2002a] found that in spring and early summer the polluted North American layers most frequently entered Europe at 5–

8 km altitude and north of 60°N . Thereafter, the North American CO tracer in general moved southward and downward toward the surface south of the Pyrenees and Alps. The present study confirms these results since it observed a strong impact of North American emissions on mountain and elevated sites in the Alpine region.

[62] One further important question is whether the effect of the long-range transport of pollution is seen in multiyear O_3 records. Brönnimann *et al.* [2002] investigated trends in near-surface O_3 concentrations in Switzerland for the 1990s. They observed a positive linear “base trend” of around $0.4 \text{ nmol mol}^{-1}$ per year which was attributed to increasing background O_3 (partly due to long-range transport). The results from our experimental studies from November 2001 show that pollution plumes transported all the way from North America can pollute the European free troposphere and mountain top sites significantly. Still, the influence from these North American pollution plumes at low altitude sites over Europe is more difficult to observe, probably due to mixing with fresh polluted boundary layer air. The CONTRACE measurements indicate that the North American pollution plume investigated here (case 19 November) was “cleaner” with respect to CO and NO_y than the average local pollution situation at low altitude sites in Central Europe. The excess O_3 observed in the plume would probably very soon decrease down to ambient mixing ratios due to mixing with boundary layer air and titration by NO, and not persist over several days like the observations in the free troposphere indicate. However, in summer the situation would be very different since during daytime photochemical O_3 production dominates over the titration by NO, and the excess O_3 from North America would be added to an already elevated ozone background. In summer, however, we also have to take into account the impact of photo-dissociation of O_3 . More field campaigns and long-

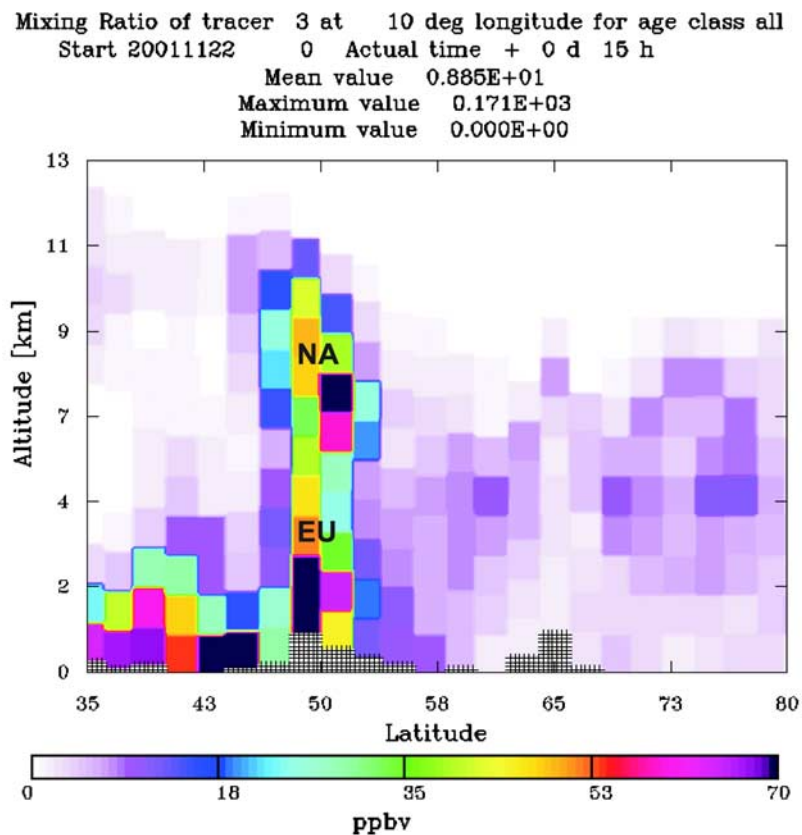


Figure 16. Vertical cross section through the FLEXPART European (EU) and North American (NA) CO tracer fields (forecast) for 22 November 2001, 1500 UTC along 10°E.

term measurements of trace gases like CO and O₃ at remote surface sites are desired to monitor this type of pollution events.

9. Summary and Conclusions

[63] Under the framework of the CONTRACE project aged North American pollution plumes passing over Europe were investigated. For the first time a Lagrangian particle dispersion model (FLEXPART) was used to predict these pollution events (CO tracer) over Europe. The aircraft measurements confirmed the presence of polluted North American air masses in several cases. One of these events (19 November 2001) was investigated here in detail. Ground measurements, MOZAIC flights, and satellite images were analyzed in the regions of the United States where the FLEXPART results suggested that the plume originated. It turned out that in mid-November 2001 the southern and eastern United States was heavily polluted, and O₃ and SO₂ were strongly enhanced. Moreover, the FLEXPART model was run to simulate the transport pathway of the pollution plume over the Atlantic and its dispersion over Europe. The passage of the NA plume over Europe took almost 3 days. A number of chemical measurements were used to identify the plume on its way across Europe. Airborne measurements identified the plume over Scandinavia (2–4 km altitude), ground sites in the Alpine region were analyzed in detail, and a Finnish ozonesonde

identified the plume before it left Europe. However, a detailed diagnosis of the chemical evolution of the NA plume over Europe was not possible from this event since the measurements were not designed as a Lagrangian experiment (repeatedly investigation of exactly the same air mass). Instead the presented measurements are a selection of random samples available along the track of the pollution plume. From the MOPITT satellite CO images are available for the pollution event showing evidence for a polluted layer leaving the U.S. east coast on 14–15 November and reaching Norway on 19 November. However, due to the rather low spatial and temporal resolution for the study of single pollution events these images were not discussed here in detail.

[64] For more detailed analyses, trace gas correlations from the airborne measurements were determined. Correlations in the NA pollution plume were compared to the unpolluted free troposphere in general. The observed NO_y/CO slope in the pollution plumes was used to estimate the time since the pollutants were emitted. The estimated time was in good agreement with the FLEXPART age spectra. Furthermore, a positive O₃-CO correlation was observed in the NA plume, which suggests that photochemical O₃ production must have been taken place. However, the season (late autumn), the northerly pathway of the polluted air mass passing Greenland and the transport in the lower-mid troposphere are factors which likely inhibited in-transit O₃ production. The observations indicate that the

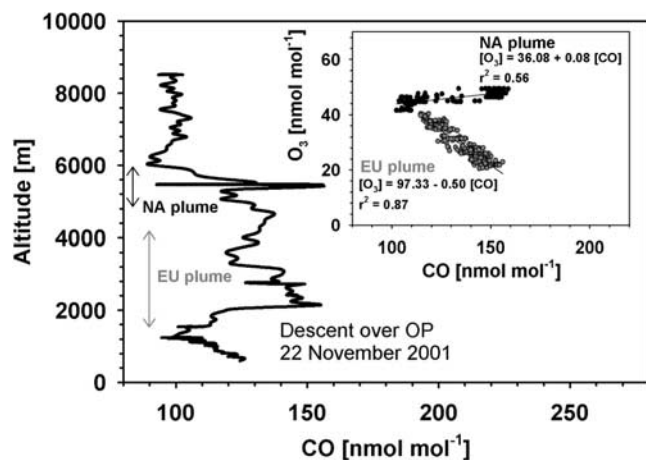


Figure 17. Measured vertical CO profile (10 s mean values) during the descent close to Oberpfaffenhofen (48°N, 11°E) on 22 November 2001. Superimposed (upper right corner) are O₃-CO correlations in the European (EU) and the North American (NA) pollution plume.

enhanced levels of ozone were already produced near the source region over the eastern United States and not during the transit.

[65] Since the pollution plume descended over the Alpine region, these European ground sites there were most strongly affected by the plume. For example at Arosa (1840 m) in the Swiss Alps ozone was elevated by 12 nmol mol⁻¹ (33%) during the two days of the plume passage in comparison to the monthly average. The airborne measurements also indicated an O₃ enhancement of ~10 nmol mol⁻¹ in the pollution plume. These experimental findings are in good agreement with model studies carried out by Li *et al.* [2002]. Especially during “events” of pollution transport from North America the surface O₃ concentration over Europe increased significantly by 5–10 nmol mol⁻¹ in their model. The impact of the NA pollution on Alpine sites was largest for mountain and elevated sites (above ~1500 m). For some lower located sites the NA pollution plume was cleaner with respect to CO and NO_y than the average local pollution situation.

[66] Finally, a second North American pollution event (22 November 2001) was briefly described where the meteorological and chemical situation was more complex than in the first case. The North American pollution plume was layered above a European pollution plume. In this case FLEXPART CO tracer forecasts and simulations were indispensable for the interpretation of the airborne measurements. The observed tracer correlations (CO-NO_y and CO-O₃) gave further information about the different chemical composition in the North American and European pollution plume.

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