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Intercontinental transport and its influence on the ozone concentrations over central Europe: Three case studies

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[1] In this paper we report on a detailed analysis of the first clear observation of ozone import from North America in the free troposphere over central Europe in May 1996 and of two more recent cases of transatlantic transport. The analysis is based on calculations with the FLEXTRA trajectory model and the FLEXPART tracer model and data from Measurements of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) flights and North American surface sites. Lidar measurements carried out under conditions of developing anticyclonicity at Garmisch-Partenkirchen, Germany, consistently yield peak ozone mixing ratios in the middle and upper troposphere between 80 and 110 ppb during the warm season. These layers are traced back to North America by FLEXTRA trajectories. The prevailing transport pathway involves a warm conveyor belt exporting high amounts of ozone, formed during a prefrontal high-ozone episode, from the central part of the eastern United States to the upper troposphere. The polluted air may enter central Europe above an airstream descending from the stratosphere, thus inverting the normal atmospheric stratification, with stratospheric air at low levels and boundary layer air in the upper troposphere. However, the first two cases studied in this paper show that the export from the eastern United States may arrive over Europe at times prior to the onset of the anticyclonic conditions. Analysis of these cases reveals the complexity of the air export from North America. The ozone maxima occurring during the observational periods are traced back to the planetary boundary layer (PBL) in other source areas in the United States. Some of them are related to entrainments even from source areas beyond North America, either in the stratosphere or in Asia. The third case is quite different in its advection pathway, with almost straight and horizontal advection at 6 to 8 km above sea level starting from the Great Lakes area. A positive correlation between ozone and aerosol in that layer verifies the presence of PBL air. However, the very high ozone mixing ratio (up to 130 ppb) and an anticorrelation of ozone and humidity suggest the entrainment of another component from long-range transport in the upper troposphere from beyond North America. In two of the case studies the export from the PBL was presumably influenced by large-scale convection. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS:* intercontinental transport, transatlantic transport, ozone, lidar, trajectories, particle dispersion model

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1. Introduction

[2] For a long time there was a substantial discrepancy between the observed export of air pollution from the North American east coast to the Atlantic Ocean [e.g., *Parrish et al.*, 1993; *Fehsenfeld et al.*, 1996a, 1996b] and the import monitored at stations in the west of the European continent [*Derwent et al.*, 1998; *Dutot et al.*, 1997; *Fenneteaux et al.*, 1999]. For many investigations by European researchers the marine sector has been regarded as clean because of background-type ozone and precursor concentrations. On

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the other hand, the United States is known to contribute significantly to the total global emissions; for example, almost one third of the "carbon" emissions [*Sandalow and Bowles*, 2001, and references therein]. *Jacob et al.* [1993] estimated that 70% of the "pollution ozone" produced over the United States is exported. Within the North Atlantic Regional Experiment (NARE) air masses leaving the eastern United States were traced forward to the central Atlantic. Elevated ozone and NO_x concentrations could be observed at 1 km above sea level (asl) on the Azores Islands [*Parrish et al.*, 1998]. Even further to the east, on the island of Tenerife, the observation of enhanced ozone in anthropogenically modified air masses led to speculations on a North American origin [*Schmitt*, 1994]. In a case study of a North American air mass arriving at Tenerife, *Schultz et al.* [1998] concluded that no significant chemical transformation of the air had taken place on its way across the ocean. During aircraft flights to the west of Europe, *Penkett et al.* [1998] found a pronounced layering of peroxides up to more than 7000 m. *Arnold et al.* [1997] detected elevated SO_2 and acetone concentrations during a flight to the northwest of Ireland at a height of 9000 m. In their NARE overview, *Fehsenfeld et al.* [1996b] concluded that long-range transport of North American air pollution takes place preferably at heights above 2.5 km, whereas ozone is efficiently destroyed in the marine boundary layer.

[3] During our high-density lidar investigations of stratospheric air intrusions within the VOTALP (Vertical Ozone Transport in the Alps) and STACCATO (Influence of Stratosphere-Troposphere Exchange in a Changing Climate on Atmospheric and Oxidation Capacity) projects we detected a richly layered structure in the ozone time series under conditions of developing anticyclonicity [*Eisele et al.*, 1999]. The peak ozone mixing ratios in the middle and upper troposphere vary between 80 and 110 ppb during the warm season.

[4] A first detailed analysis was carried out for an episode on 28 and 29 May 1997, which was particularly easy to interpret since the ozone maxima could be attributed to air import from the boundary layer in a single source region located in the eastern United States [*Stohl and Trickl*, 1999]. Transport modeling with the FLEXTRA and FLEXPART models showed that the detected ozone-rich boundary-layer (PBL) air left the North American coast to the southeast where it was lifted to about 10 km in a warm conveyor belt (WCB). Subsequently, rapid transport within the jet stream and anticyclonic subsidence took these layers to the observational area in southern Germany. Surface data for the eastern United States and a MOZAIC (Measurements of Ozone by Airbus In-Service Aircraft [*Marenco et al.*, 1998]) flight through the WCB confirmed the ozone values of the order of 90 ppb found by the lidar measurements at IFU.

[5] In contrast to the North American export cases investigated within NARE the ozone-rich air masses that eventually reached central Europe in the middle and upper troposphere left the east coast of the United States at lower latitudes. The export took place to the southeast and the overall transport path had a wave-like pattern, resembling an S rotated by roughly 90° and the central part of the S being formed by the WCB. Such transport occurs quite frequently over the North Atlantic, whose dimensions are comparable to the wavelength of baroclinic waves developing at the polar front. Thus WCBs often take up PBL pollution over the eastern United States, at the beginning of the North Atlantic storm track, transport it across the ocean, and deposit it in the upper troposphere over Europe, near the end of the storm track. In contrast, because of its larger dimensions, the North Pacific storm track actually consists of at least two subsets [*Hoskins and Hodges*, 2002], and thus transport over the Pacific typically involves transport around several troughs and ridges, which leads to stronger dilution of polluted plumes. The trace gas composition in the rising and descending airstreams of midlatitude cyclones over the western North Atlantic was established by *Cooper et al.* [2002a, 2002b].

[6] The high frequency of transatlantic transport episodes and the transfer of large volumes of polluted air during them [*Stohl et al.*, 2002a] underline their potential importance for the European free-tropospheric ozone background. In this paper, we give a detailed analysis of three more of the (at least) seven cases for which high-ozone input from North America has been observed within VOTALP and STACCATO [see also *Roelofs et al.*, 2003; *Zanis et al.*, 2003], which demonstrate that ozone import from across the Atlantic is substantially more complex than previously thought. These cases were each characterized by peak ozone values in the middle and upper troposphere consistently higher than 80 ppb, over periods up to five days. The methods and data used are described in section 2, the three case studies are investigated in detail in section 3 and conclusions are drawn in section 4.

2. Methods

2.1. Lidar Measurements

[7] The stationary ozone lidar of IFU [*Eisele and Trickl*, 1997], located at Garmisch-Partenkirchen (Bavarian Alps, 47°28′37″N, 11°03′52″E, and 740 m asl), is a three-wavelength differential-absorption system operated at 277 nm and 292 nm as the "on" wavelengths and 313 nm as the "off" wavelength. The high light absorption by ozone at 277 nm allows us to retrieve vertical profiles of ozone with high accuracy (3 to 5 ppb) and a vertical resolution of 50 to 200 m up to about 7 km, the 292-nm channel extends the vertical range into the lower stratosphere (3 to 5 km above the tropopause), with a vertical resolution of the order of 0.5 km. Because of the lower sensitivity of this channel the vertical resolution for the ozone retrieval is usually chosen lower by a factor of two with respect to 277 nm. The accuracy achievable in the upper troposphere is approximately 5 ppb, but the upper tropospheric results may be degraded if there is strong light absorption in the presence of elevated ozone. A single measurement takes 45 s. During periods of intensive vertical sounding measurements are carried out under automatic control at intervals of 1 h. All time series shown in this paper are given in Central European Time (CET = UTC + 1 h). Most observations of intercontinental transport with our ozone lidar have so far been associated with the onset of anticyclonic conditions, when subsidence leads to clear conditions, which is ideal for optical sounding [*Eisele et al.*, 1999].

[8] For aerosol studies in the free troposphere, the stratospheric aerosol lidar of IFU [e.g., *Freudenthaler et al.*, 1994; *Jäger et al.*, 1997] was used in one case. This lidar

yields data for heights above 3 km asl and, because of its applications in the tropopause region and the stratosphere, has a much better sensitivity in the free troposphere than the tropospheric aerosol lidars of the institute. The sensitivity for aerosol at its operating wavelength of 532 nm is substantially higher than that of the ozone lidar (313 nm) because of the much lower signal background from Rayleigh backscattering.

2.2. Transport Models

[9] Two models were used to relate the measurements to possible source regions: First, backward trajectories ending in an array along the lidar profile for heights up to 12 km asl at vertical intervals of 0.25 km and time intervals of 3 h were calculated using the FLEXTRA trajectory model [*Stohl et al.*, 1995; *Baumann and Stohl*, 1997; *Stohl and Seibert*, 1998]. FLEXTRA is driven with global analysis fields every 6 h and with 3-h forecast fields every other 3 h. The wind fields have a resolution of 1° both in longitude and latitude and were retrieved from the *European Center for Medium-Range Weather Forecasts* (*ECMWF*) [1995] archives. FLEXTRA uses bicubic horizontal, quadratic vertical and linear time interpolation. No isentropic assumption is invoked, which is important especially for ascending airstreams where condensation causes diabatic heating.

[10] The second model used is the Lagrangian particle dispersion model FLEXPART (version 4.0), which was also driven with ECMWF data [*Stohl et al.*, 1998; *Stohl and Thomson*, 1999]. FLEXPART calculates the transport and dispersion of non-reactive tracers. It has been validated with data from three large-scale tracer experiments in North America and Europe [*Stohl et al.*, 1998] and performed very well in comparison with other models. *Forster et al.* [2001] used FLEXPART to study the transport of pollutant plumes from Canadian forest fires to Europe. FLEXPART treats advection and turbulent diffusion by calculating the trajectories of a multitude of particles. Stochastic fluctuations of the three wind components, obtained by solving Langevin equations [*Stohl and Thomson*, 1999], are superimposed on the grid-scale winds interpolated from ECMWF data to simulate transport by turbulent eddies. In the PBL, the magnitude and the Lagrangian decorrelation times of the turbulent wind components are obtained from a parameterization by *Hanna* [1982]. Above the PBL, the turbulent components are set to small values in dependence of the wind shear. Recently, the convection scheme of *Emanuel and Zivkovic-Rothman* [1999] was implemented in FLEXPART. Documentation manuals and the source codes of both FLEXTRA and FLEXPART can be obtained via the Internet from the address http://www.fw.tum.de/EXT/LST/METEO/stohl/.

[11] FLEXPART was used to advect two passive tracers: an NO_x emission tracer and a tracer for stratospheric ozone. The NO_x tracer was released over North America in order to study whether air masses observed by the lidar were possibly contaminated by North American emissions. Model tracer releases were typically started about two weeks before the onset of the measurements. The emission distribution was taken from the GEIA emission inventory for the year 1985 [*Benkovitz et al.*, 1996]. Typically, 10 million particles were released in 1° × 1° grid cells at a constant rate during a simulation, with the number of particles released within a grid cell scaled with the emission strength of that cell. All particles carried the same mass, and as no removal processes were considered, the mixing ratios reported over Garmisch-Partenkirchen indicate the total North American NO_x emissions into the observed air masses during the period considered.

[12] For the stratospheric ozone tracer, the model setup was similar to the one described by *Stohl et al.* [2000] and *Stohl and Trickl* [1999], but with an extended model domain stretching from 80°W (130°W in one case) to 40°E and from 25°N to 82°N and up to 16 km above ground level. This domain was homogeneously filled with particles at the start of a simulation. Only stratospheric particles were considered; tropospheric ones were removed from the simulation during initialization. A dynamical definition of the tropopause, based on a threshold value for the potential vorticity (PV) of 1.6 potential vorticity units (pvu, 1 pvu = 1×10^{-6} m^2 K kg^{-1} s^{-1}), is used. During a simulation, particles are created in the stratosphere at the inflowing boundary, and are destroyed at the outflowing boundary. Approximately 4.5 million particles were present at any time in the model domain. Each particle carries a certain amount of ozone so that initial ozone concentrations follow the relationship with PV: O_3 [ppb] = S [ppb/pvu] × PV [pvu] [*Danielsen*, 1968; *Beekmann et al.*, 1994], with S being highest in March and lowest in October [*Stohl et al.*, 2000]. Ozone is a passive tracer in the model, but is subject to dry deposition at the surface.

[13] In some of the cases for which the trajectories, because of their strong decrease in density over North America and neglect of turbulence, did not yield clear information on the source regions, backward tracer calculations with FLEXPART were performed which results in a much better statistical performance [*Stohl et al.*, 2003]. For these calculations tracers were released above Garmisch-Partenkirchen in a small 3-D box over a narrow time window. Residence times in the total atmospheric column and at an altitude of 500 m above the ground were derived. Furthermore, the integral of all tracers residing in this surface layer was computed for up to two weeks backward in time, an additional option being folding with the data from the 1995 EDGAR emission inventory in order to visualize the most likely source area.

2.3. MOZAIC Measurements

[14] Measurements of ozone and water vapor are carried out onboard five commercial airliners within the MOZAIC (Measurements of Ozone and Water Vapor by Airbus In-Service Aircraft) program [*Marenco et al.*, 1998]. Data from about eight flights per day are available in the MOZAIC database, with most of the flights being directed from Europe to destinations in North America. This data set, providing daily vertical profiles over Europe, transects at flight level over the North Atlantic and occasional vertical profiles at various locations over North America, and is highly valuable for the studies presented here. Data from a large number of flights have been examined. The data resolution is one minute or less, and vertical resolution is 150 m or better.

2.4. GOES-East Water Vapor Satellite Images

[15] This study utilizes water vapor imagery from the National Oceanic and Atmospheric Administration's

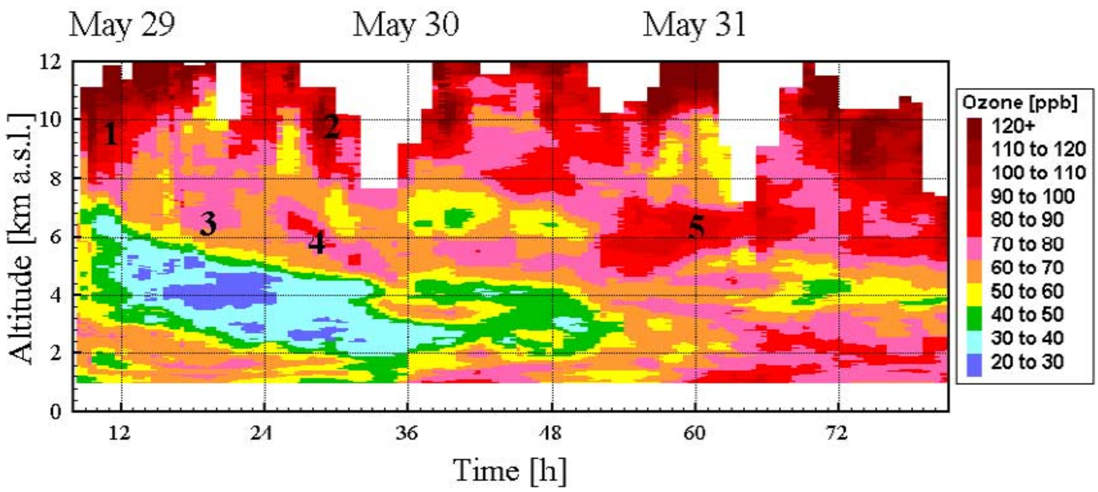

Figure 1. Four-day series of ozone soundings with the IFU ozone lidar (29 May to 1 June 1996); for details, see text. The time is given in Central European Time (CET = UTC + 1 h).

(NOAA) Geostationary Operational Environmental Satellite centered above the equator and 75°W, known as GOES 8 or GOES-East. The images depict radiation emitted at the 6.7 μm wavelength. Radiation at this infrared wavelength is emitted by water vapor in the mid and upper troposphere, with water vapor at warmer temperatures emitting greater amounts of radiation. These water vapor images can be interpreted as relative humidity in the mid to upper troposphere [*Soden and Bretherton*, 1993; *Moody et al.*, 1999], with the blues and purples indicating relatively dry air and the greens, yellows and reds indicating relatively moist air.

2.5. Surface Data From the United States

[16] Surface data from the United States was derived from the AIRS graphics of the U.S. Environmental Protection Agency (PSI Chart (Pollutant Standards Index), http://www.epa.gov/agweb/). For ozone, a PSI value of 100 corresponds to a maximum hourly average in a given district of 120 ppb or an eight-hour average of 80 ppb (http://www.epa.gov/ttn/amtic/psi.html), no longer available). We converted the PSIs to mixing ratios on an hourly basis. This approach is not fully quantitative, but the PSI time series have allowed us to identify high-ozone episodes in the regions of interest. In some cases the approximate correctness of our conversion could be verified by MOZAIC measurements.

[17] For the years after 1997 ozone maps (hourly averages) for some parts of the United States were copied from http://www.epa.gov/airnow/archives/. These maps mostly cover areas with sufficiently high density of monitoring stations.

3. Results

3.1. Case 1: 29 May to 1 June 1996

[18] Figure 1 is a revised version of Figure 4 of *Eisele et al.* [1999] and displays the first four-day series of ozone soundings with the upgraded IFU lidar during the passage of a high-pressure zone across central Europe. The figure shows several subsiding layers of elevated and low ozone as well as the build-up of PBL ozone starting on 30 May. On 29 May the lowest of the layers (below 3.5 km asl) is caused by a stratospheric air intrusion reaching the ground. This intrusion has been well characterized in a number of modeling studies and observations at mountain sites [e.g., *Feldmann et al.*, 1999; *Stohl et al.*, 2000; *Cristofanelli et al.*, 2003; *Meloen et al.*, 2003]. During the first two days of observation a low-ozone layer 2 km wide was observed above the intrusion. The ozone mixing ratio was in part as low as 25 ppb and could be explained by advection from the central Atlantic [*Eisele et al.*, 1999]. Following the intrusion period, the height range up to 4 km was characterized by anticyclonic conditions and the formation of an Alpine PBL [*Carnuth and Trickl*, 2000; *Carnuth et al.*, 2002], exhibiting elevated aerosol and ozone mixing ratios in part exceeding 80 ppb (which is rather high for the Bavarian Alps). In the middle and upper troposphere, peak ozone mixing ratios of 80 to more than 100 ppb persisted over the full observational period of four days, which was not immediately understood. As we know from the model calculations cited above, these puzzling structures cannot be explained by stratospheric air intrusions over the eastern half of the North Atlantic or Europe. The results of our rather detailed analysis of these layers are described in the following. The measurements ended in the afternoon of 1 June because of the onset of thunderstorms.

[19] FLEXTRA backward trajectories show that the air masses arriving above the subtropical layer have spent some time over the United States. As two examples we show in Figures 2 and 3 FLEXTRA backward trajectory results for 29 May (9 UTC) and 31 May (18 UTC), respectively, selected from two important periods of the measurements. The transport pathway resembles that for the May 1997 case discussed by *Stohl and Trickl* [1999] and mentioned in the introduction. Again, the air eventually reaching Garmisch-Partenkirchen left the eastern United States in a southeasterly direction and is partly entrained into a WCB over the Atlantic. However, the transport is considerably more com-

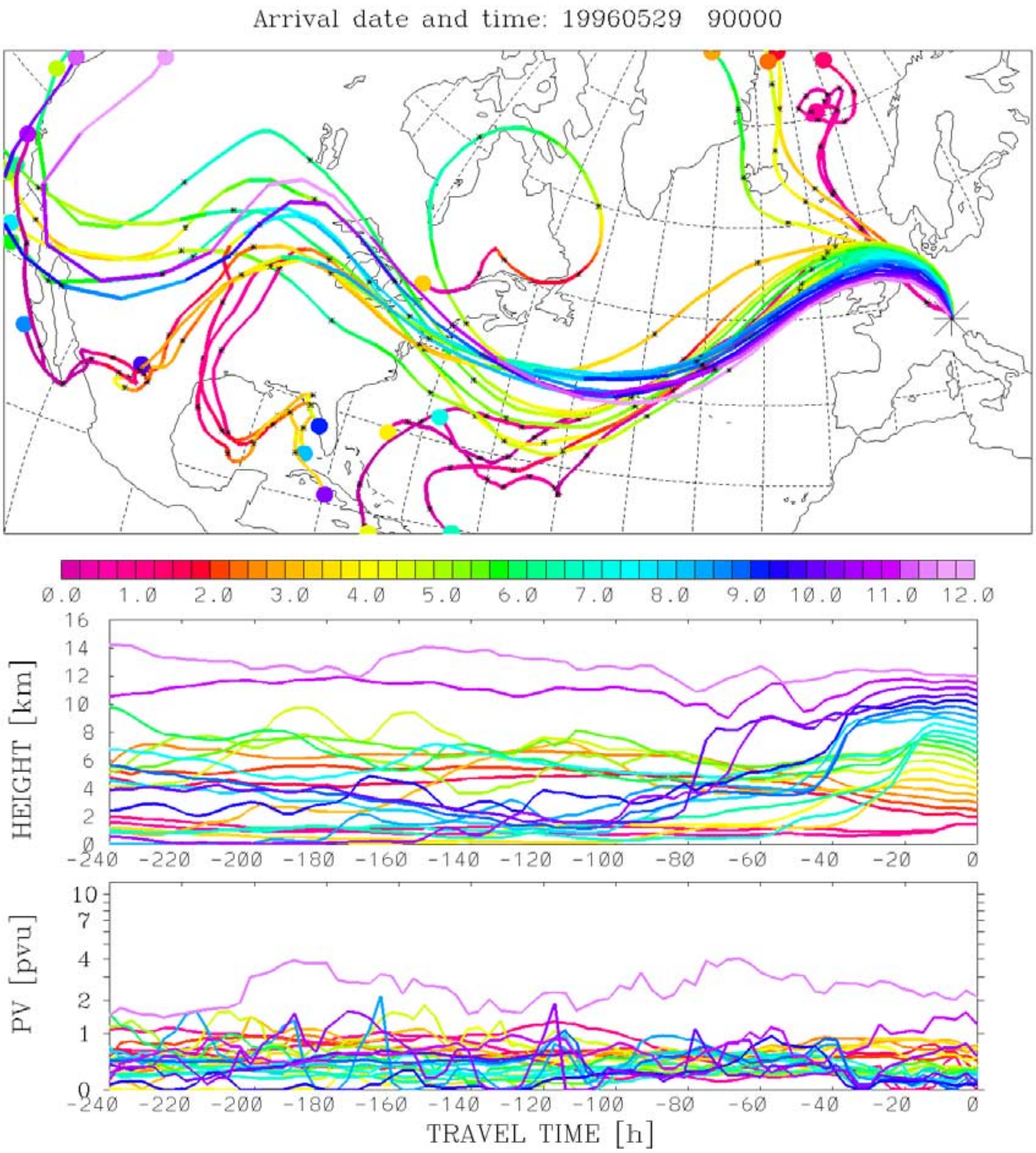

Figure 2. FLEXTRA backward trajectories for Garmisch-Partenkirchen (29 May 1996; 9:00 UTC); the height is color-coded. In the upper panel the color directly shows the actual height in kilometers. The color of the filled circles at -10 d denotes the height of the respective trajectory over Garmisch-Partenkirchen, which facilitates the assignment. In those cases in which a trajectory leaves the map within ten days, these dots were placed on or next to its intersection with the frame. One-day intervals are marked on the trajectories by black crosses. In the middle and lower panels the color is chosen according to the arrival height.

plex than in the 1997 textbook example, and many different air masses reach the lidar site during the four-day measurement period. In contrast to the May 1997 episode, just a few trajectories lead back to the PBL of the eastern United States within the ten-day calculation period.

[20] The middle and upper tropospheric observations shown in Figure 1 may be divided into two halves, which are discussed in some detail in the following. In addition, the question of the low import from the PBL of the eastern United States during the observational period is addressed. The most important time-height zones for the analysis described below are labeled in Figure 1 with numbers (1 to 5).

3.1.1. Period 29 and 30 May

[21] Figure 2 describes the transport situation shortly after the beginning of the lidar measurements. Trajectories arriving at the lowest levels (0 to 3 km) originate from the northwest. Those passing over Iceland descend strongly and are associated with the stratospheric intrusion. Trajectories arriving between about 3 and 4 km show a strong shear from a northwesterly to a southwesterly origin, with those at higher levels, up to about 6 to 7 km ascending from the

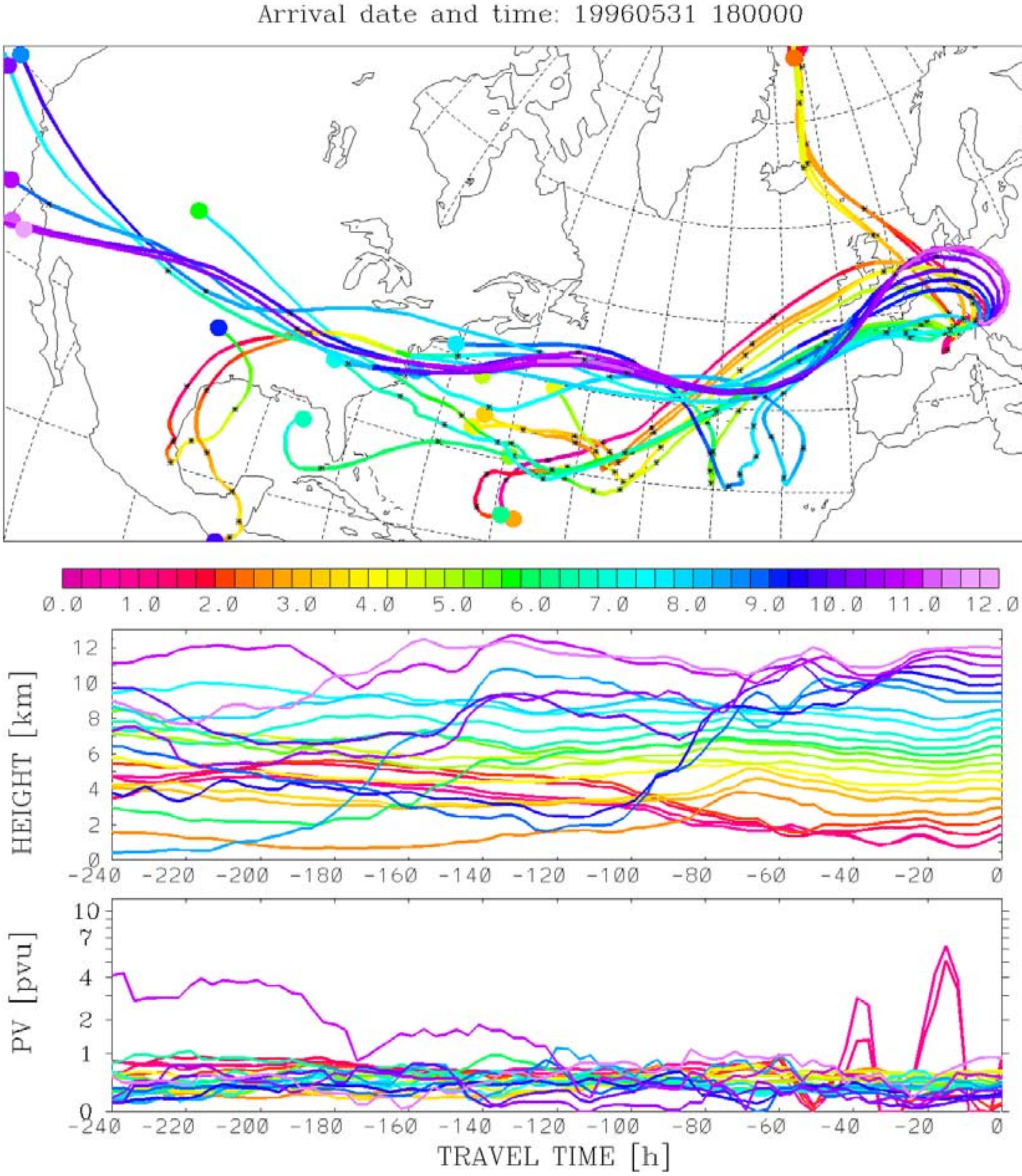

Figure 3. FLEXTRA backward trajectories for Garmisch-Partenkirchen (31 May 1996; 18:00 UTC); for details, see Figure 2.

(sub)tropical Atlantic. These are associated with the low ozone concentrations seen in Figure 1.

[22] Above about 7 km, the trajectories have a more northerly origin, once again. They may be divided into two types indicated by the two steps in the central panel of Figure 2 at −20 h (6 to 8 km) and −40 h (8 to 11 km). At later arrival times the type-one trajectories correspond in Figure 1 to the subsiding part of the high-ozone region just above the subtropical air mass. This layer corresponds to complex input from different source regions during the two periods marked by labels 3 and 4 in Figure 1. Since the descending air mass contains some import from the eastern United States on 29 May (label 3) we give the details on the periods around labels 3 and 4 in the chapter on the ozone import from the eastern United States further below. The type-two trajectories always stay in the uppermost troposphere during the final air mass approach to the lidar site, associated with rapid travel with the jet stream. They show a strong ascent from the North American PBL (altitudes below 1 km are reached over Texas and western and central Mexico) associated with a WCB starting between −75 and −100 h, to the south west of the Great Lakes. These trajectories arrive in a layer of very high ozone mixing ratios in part exceeding 100 ppb (next to labels 1 and 2 in Figure 1). Both bundles pass close to major urban source regions of ozone precursors (e.g., Houston and Mexico City, which are well known as being among the cities with the most severe ozone episodes over North America). The maximum PSI values for Texas were moderate during the second half of May, but more than 70 downwind of Houston for the period between 21 and 23 May, however somewhat dropping afterward. According to the trajectory

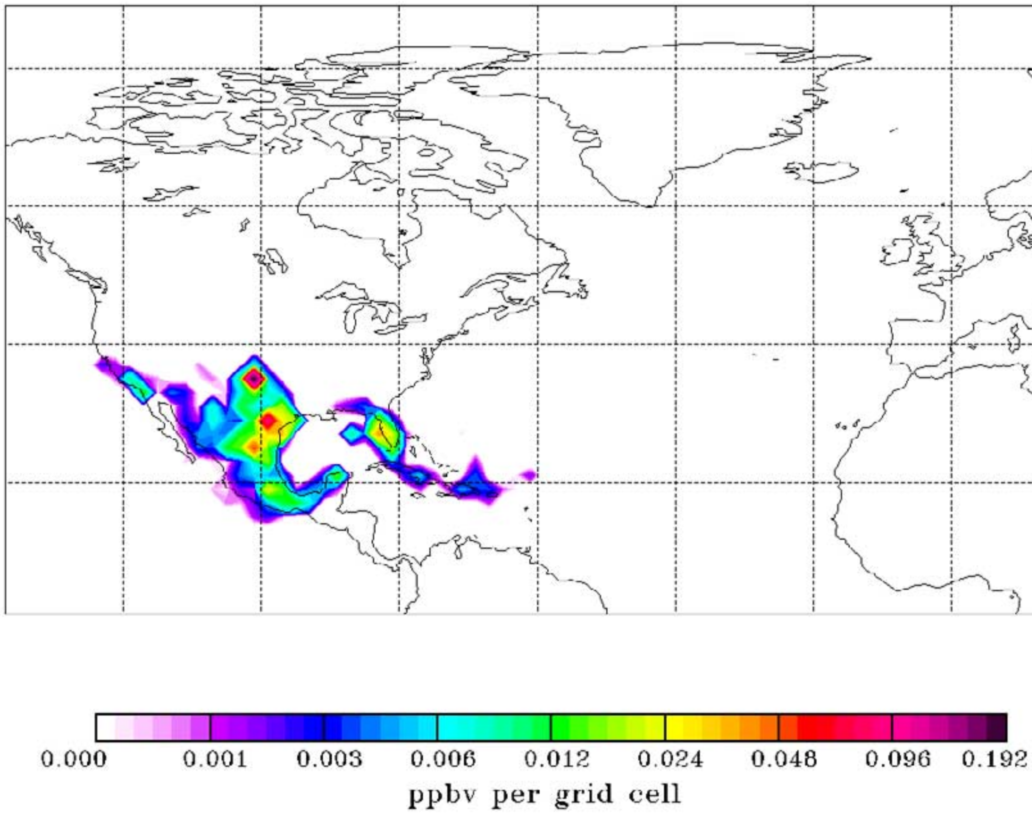

Figure 4. Backward tracer simulation for 30 May 1996, 3 to 6 UTC, initiated over Garmisch-Partenkirchen at altitudes between 9 and 11 km asl and showing the relative importance of the different source areas in North America (Amarillo, Houston, Monterrey, and, to some extent, Mexico City) based on weighting with a priori emissions.

calculations, transport from the North American PBL to Garmisch-Partenkirchen becomes somewhat less important for arrival times toward the end of 30 May. The import from Mexico was most important on 30 May. Because of the low density of the trajectories in these remote areas an additional backward tracer run was carried out, with particles initialized over Garmisch-Partenkirchen in the time interval between 3 and 6 UTC and at heights between 9 and 11 km (marked in Figure 1 by label 2). The integrated result for 500 m above the surface, folded with the emissions from the EDGAR inventory, is shown in Figure 4 and confirms the input from a major part of Mexico including Mexico City, in addition to that from Texas.

[23] Figure 5 shows ozone and relative humidity profiles for a MOZAIC departure from Frankfurt at about 8:15 UTC (9:15 CET). Because of the difference in air mass arrival time over Garmisch-Partenkirchen these profiles correspond to the early afternoon measurements with the lidar. High humidity, associated with ozone as low as 25 ppb, is seen in Figure 4 below 5.5 km (subtropical air below 4 km above Frankfurt, below 5.5 km above Garmisch-Partenkirchen) and above 8 km. The elevated-humidity ranges between 4 and 5.5.km and above 8 km correspond to the type-one and type-two trajectories, respectively. Very interestingly, both the lidar and MOZAIC measurements reveal moderate ozone values around 60 ppb above the subtropical air layer. During that short moderate-ozone phase there are almost no trajectories from Texas or Mexico. In general, there seems to be some correlation of the number of trajectories passing over that area and the measured ozone values in the upper troposphere.

[24] The GOES-East image in Figure 6 (25 May, 0:15 UTC) shows the situation at the beginning of the export of the air masses from Texas. Around this time a new front formed at 100°W (below 42°N), which was quasi-stationary for about a full day. 0:15 UTC corresponds to the late afternoon in that area which explains the formation of a mesoscale convective complex along the front, with additional thunderstorms further south. The images for the first hours of 26 May (in UTC), the period corresponding to the first air mass ascent, indicate the revival of large-scale convection north of 33°N. Because of the presence of these convective cells some entrainment of stratospheric ozone or photochemical ozone production due to NO_x formation by lightning cannot be excluded. Because of the strong convection the trajectory results could be quite uncertain, but as some of them show strong ascent in and around this convective zone, they seem to capture the basic features.

[25] Figure 7 shows the situation on 27 May, 0:15 UTC, when most of the air reaching the lidar on 29 May was over the central part of the Atlantic between America and Europe. Several fronts are seen, indicated by elevated moisture. A comparison with Figure 2 shows that the transport path during the final phase was in the vicinity of

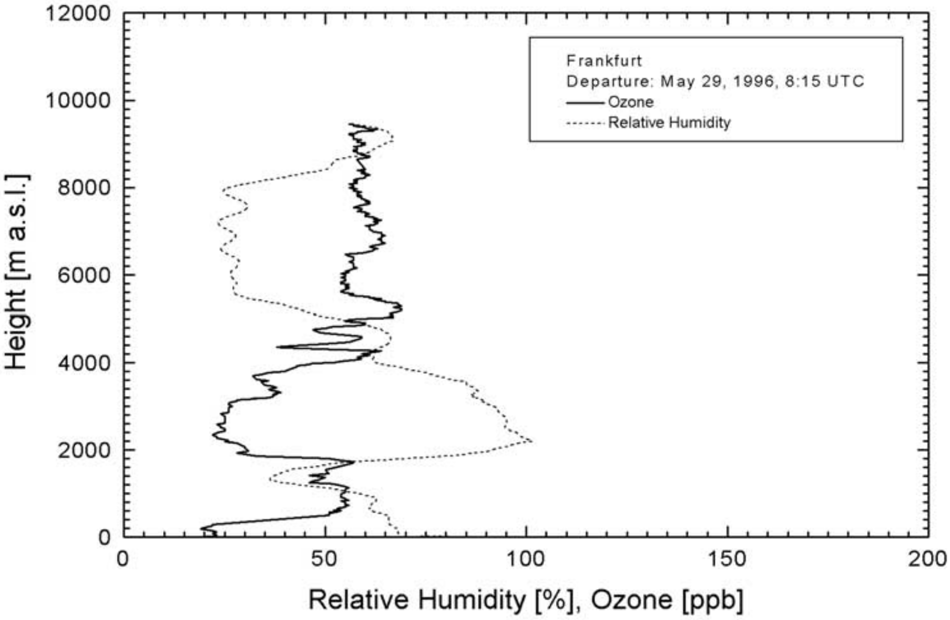

Figure 5. MOZAIC ozone and relative humidity profiles, Frankfurt, 29 May 1996, departure time roughly 8:15; the profile is (apart from some differences between 4 and 5.5 km) in nice agreement with the lidar measurements at Garmisch-Partenkirchen in the early afternoon, which is ascribed to the time difference in air mass arrival. The elevated-humidity layer above 8 km corresponds to the type-one trajectories.

the elongated frontal system between Cuba and roughly 50°N, 20°W.

[26] The MOZAIC flights between North America and Frankfurt show a pronounced departure from stratospheric air characteristics starting on 26 May, when the WCB outflow region of the main front in Figures 6 and 7 had proceeded far enough to the east to intersect the lower latitude section of the flights over the eastern Atlantic. In Figures 8 and 9 we give examples for ozone and water vapor results during two flights (Frankfurt−Dallas and Los Angeles−Frankfurt) at different flight altitudes over the eastern Atlantic. In both cases rather high relative humidity is seen to the northwest of Europe, in the region of the WCB outflow according to the trajectory results. The case for height levels near 11 km (Figure 8) shows ozone mixing ratios of the order of 120 ppb in the high-humidity part of the flight. Although the high humidity indicates the presence of a significant amount of lower tropospheric air, an exclusive PBL origin of this ozone layer cannot be concluded since there is little correlation between humidity and ozone. The flight level at 11.3 km is at the upper edge of the WCB (compare Figure 2) and entrainment of stratospheric ozone is likely.

[27] For cruising altitudes below 9 km low ozone (30 to 40 ppb) was registered between 0.5 and 8 degrees west and 70 to 90 ppb in the remaining part of the high-humidity zone in the east of 12°W (Figure 9). These mixing ratios agree with the idea of co-existing subtropical and North American air both caught in the WCB.

[28] The overall time-height distribution of the import from the North American PBL above the lidar site is synthesized by a NO_x tracer simulation with FLEXPART (Figure 10). Figure 10 shows the highest concentrations over IFU for the early phase of the observations, as expected from the FLEXTRA results, and in rough agreement with the elevated ozone concentrations observed above 6 km on 29 May. The model shows that air from the North American PBL is contained in both the upper tropospheric and the descending middle tropospheric branch already deduced from the trajectories. Some NO_x tracer is also caught in the subtropical air mass with very low ozone concentrations. However, the tracer concentrations are rather low, and the tracer has spent considerable time in the marine boundary layer. Therefore it is likely that even if ozone had been formed from the NO_x emissions, it was depleted later over the surface of the ocean. This emphasizes that in order for significant intercontinental ozone transport to occur, lifting to the middle or upper troposphere must occur close to the pollution sources. During the later period of the lidar measurements minor tracer contributions are seen at altitudes around 8.5 km and below 5 km. This suggests low input from the North American PBL at later times.

3.1.2. Period 31 May and 1 June

[29] In the second half of the observational period the humidity in the free troposphere was frequently low. Since the maximum ozone concentrations are high and since the FLEXTRA results in Figure 10 between 4 and 8.5 km do not show input from the North American PBL a more and more stratospheric nature of the air might be concluded. Therefore some calculations with stratospheric tracer were made. Figure 11 shows a FLEXPART simulation for stratospheric tracer initialized over the North Atlantic between 80°W and 40°E, thus including a larger area than the earlier modeling studies for this period. Some stratospheric contribution is seen on 31 May above 4 km, but the ozone values

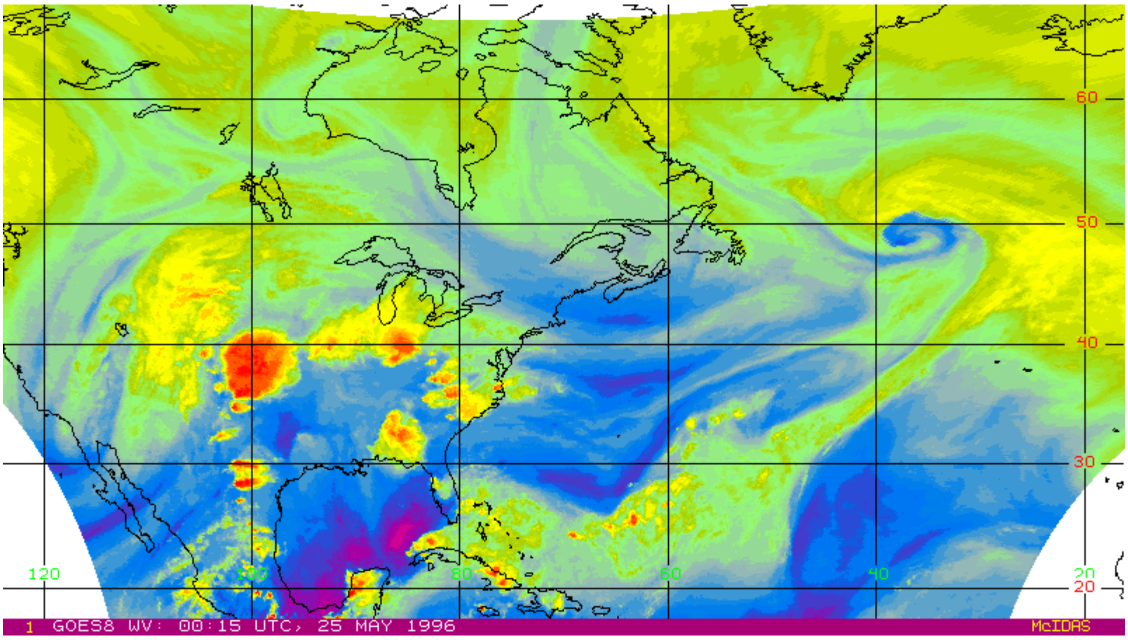

Figure 6. GOES-East water vapor image for 25 May 1996, 0:15 UTC; for technical details, see section 2.4.

are substantially lower than the 100 ppb registered by the lidar. The simulation was subsequently extended to 130°W (beyond the North American west coast), but did not reveal significant additional ozone input. Consequently, if there was any important stratospheric contribution it could only have come from still further west again.

[30] The most important layer with high ozone concentrations during the second half of the measurements was observed on 31 May between about 5.5 and 7 km asl (hours 53 to 70, see label 5 in Figure 1). This layer is confirmed by a sonde ascent at Hohenpeißenberg [*Eisele and Trickl*, 1997; *Eisele et al.*, 1997] and cannot be explained by the trajectories (Figure 3), which do not indicate an obvious source region over the previous ten days. Rather they stay in the middle and upper troposphere, and the source area may be even beyond North America. This extended layer is found in all MOZAIC profiles for Frankfurt on 31 May (an example is given in Figure 12), the lower boundary being above 4 km, in good agreement with the lidar. The relative humidity in the layer is roughly

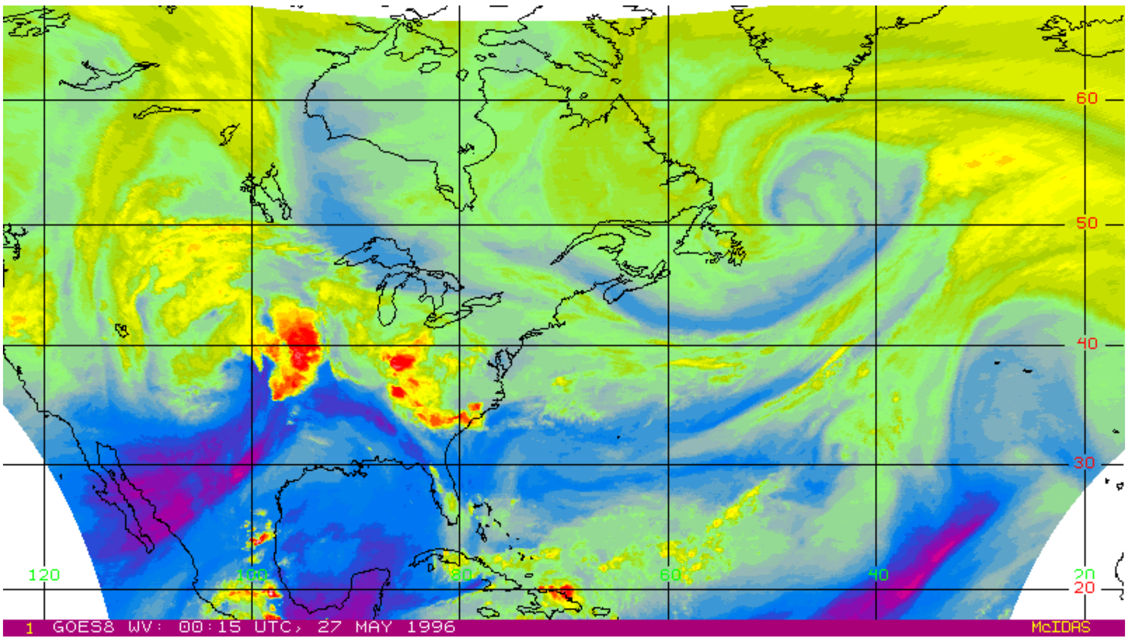

Figure 7. GOES-East water vapor image for 27 May 1996, 0:15 UTC; for technical details, see section 2.4.

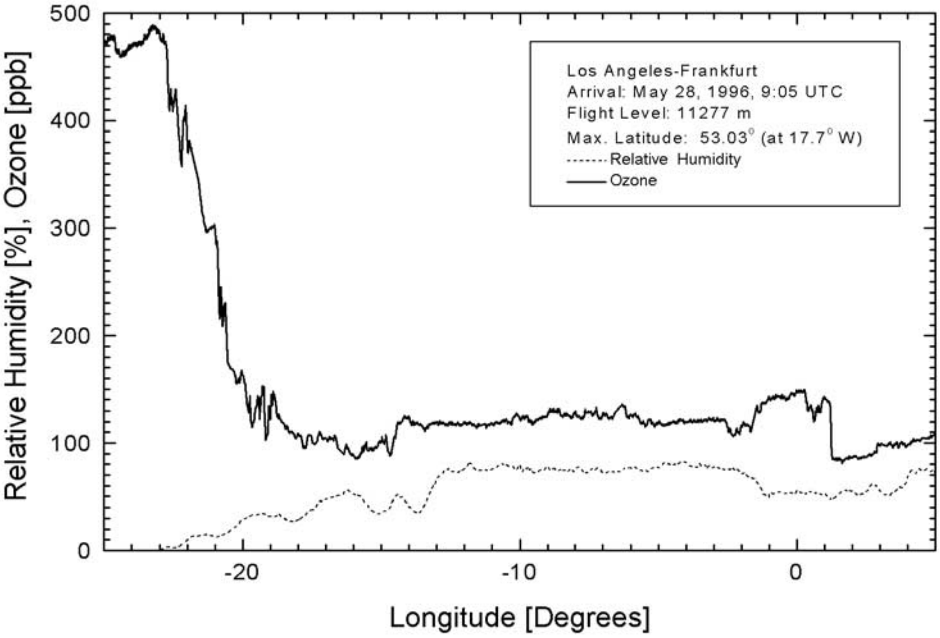

Figure 8. MOZAIC ozone and relative humidity values at a cruising altitude near 11 km, flight Los Angeles to Frankfurt, arrival: 28 May 1996, 9:05 UTC.

10% and, together with the elevated ozone values, could suggest the presence of aged stratospheric air. The PV along the backward trajectories ending in this layer is below the threshold for stratospheric air, but as PV is not conserved over such a long time, this does not exclude a stratospheric origin at earlier times. Another explanation could be that it is an aged pollutant plume from Asia. However, at present we have no means to distinguish between these two possibilities.

[31] Comparably dry layers covering the entire middle and upper troposphere are also seen in MOZAIC profiles obtained over the United States. Some MOZAIC profiles for Los Angeles (25 May) and New York (28 May) crudely fit to the passage times of the rapidly traveling upper tropospheric air mass arriving over central Europe on 31 May. In both cases low relative humidity (approximately 10% and 35%, respectively) and ozone mixing ratios exceeding 110 ppb are seen above 5 km. Los Angeles is

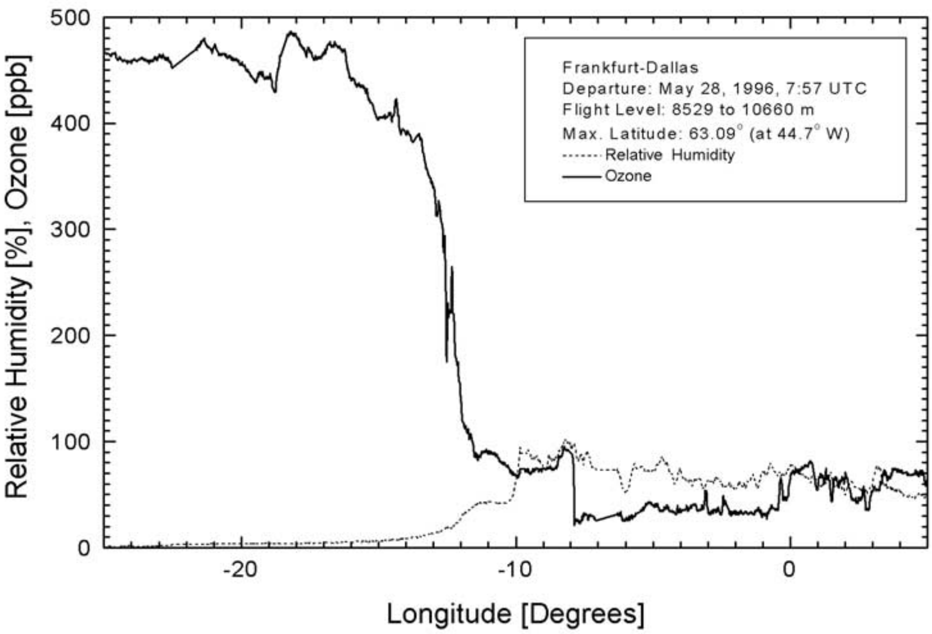

Figure 9. MOZAIC ozone and relative humidity values at an initial cruising altitude below 9 km, flight Frankfurt to Dallas, departure: 28 May 1996, 7:57 UTC.

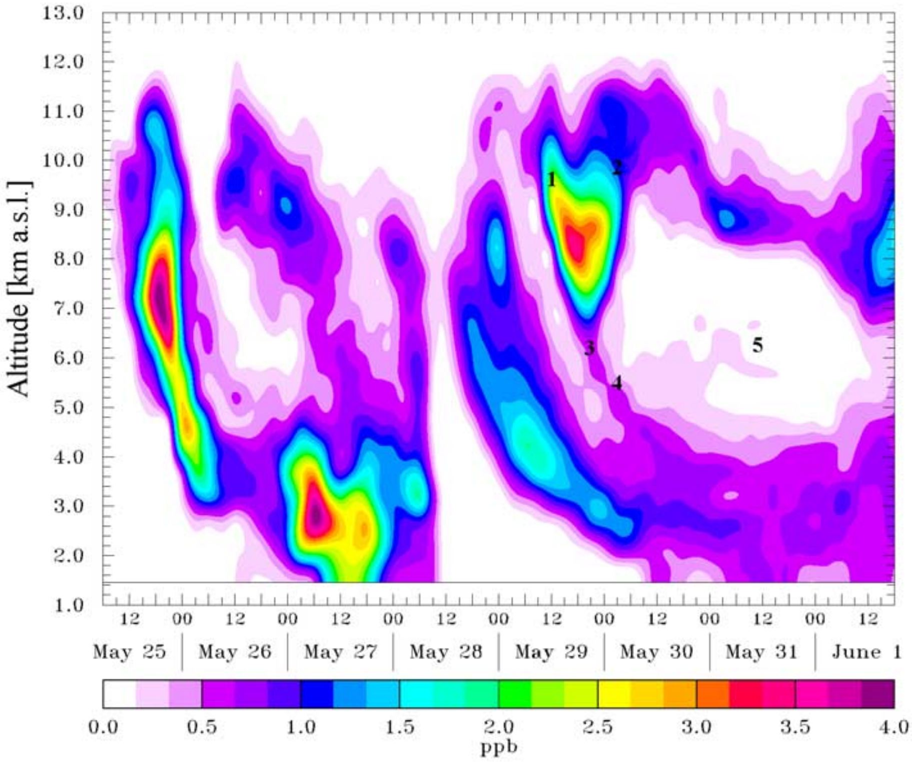

Figure 10. Section of a FLEXPART North American NO_x tracer simulation for air mass arrival above Garmisch-Partenkirchen between 25 May and 1 June 1996 (full simulation period 21 May to 3 June). The numbers correspond to those in Figure 1.

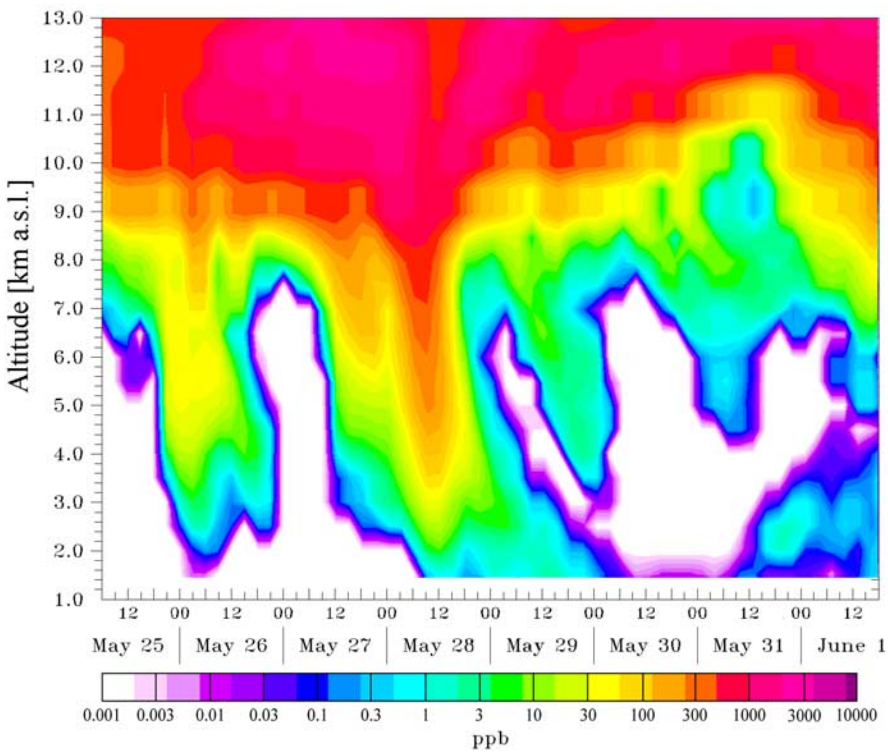

Figure 11. Section of a FLEXPART stratospheric ozone tracer simulation (25 May to 1 June) initialized above the North Atlantic (to 80° W); please, note the logarithmic mixing-ratio scale.

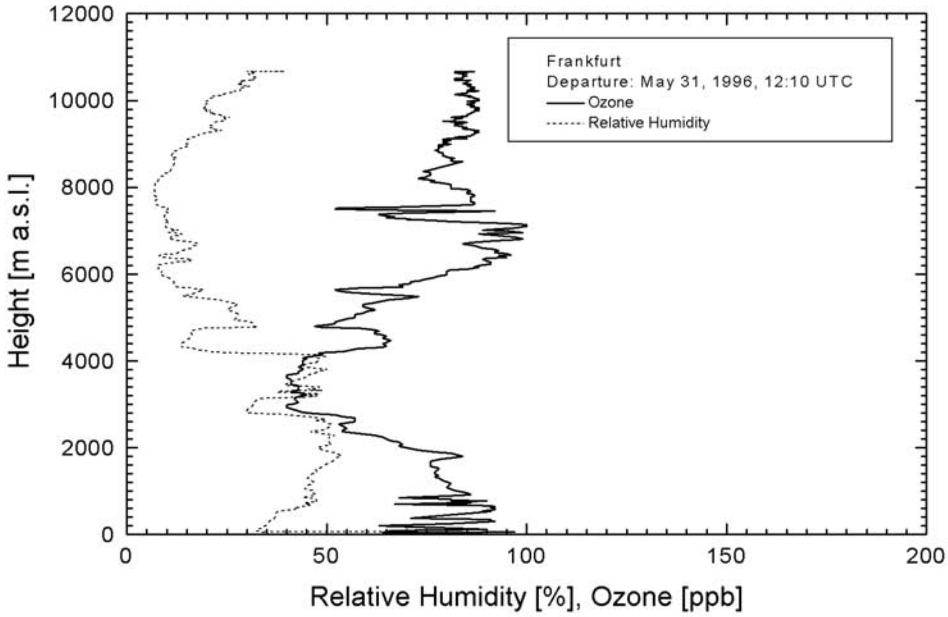

Figure 12. MOZAIC ozone and relative humidity profiles, Frankfurt, 31 May 1996, 12:10 UTC.

within the envelope of the FLEXTRA trajectories whereas New York is slightly outside the main flow in the upper troposphere, which could explain the difference in relative humidity. However, some uncertainty remains, since the trajectories arriving from the Pacific within ten days are confined to heights above 7 km. Also the GOES-East images have not allowed us to visualize an upper tropospheric airstream because of the presence of various frontal systems underneath.

3.1.3. Ozone Import From the Eastern United States

[32] The May 1997 episode [*Stohl and Trickl*, 1999] and subsequent model simulations [*Stohl et al.*, 2002a] have identified the export of air pollution from the eastern United States in WCBs as an important potential source of free-tropospheric air pollution over Europe. However, for our observations between 29 May and 1 June 1996, there is little influence from this source, which is confirmed by surface and MOZAIC data. A pronounced ozone episode was found at the central east coast during the period 19–22 May 1996, which is too early to explain any of the observations above IFU. The PSI values ranged between 70 and 93, indicating ozone mixing ratios of 84 to 112 ppb reached for at least one hour. These values are confirmed within the PBL by MOZAIC flights to/from New York. Afterward, because of a cold-front passage from the northwest to the southwest on 22 May the ozone mixing ratio dropped to about 70 ppb and to much lower values after a second frontal passage on 24 May. The air export for the days after 24 May are therefore not likely to contribute to the elevated concentrations above the IFU lidar. Subsequently, the first (larger) frontal system, which had passed the coast on 22 May, started to stretch toward Europe which is nicely visualized by the GOES-East images (Figures 6 and 7) and verified by the MOZAIC flights after 25 May (e.g., Figures 8 and 9).

[33] The FLEXPART simulation depicted in Figure 10 shows important NO_x tracer import from North America prior to the beginning of the lidar measurements. Backward tracer simulations starting at the lidar position confirm that a major part of the corresponding air mass may be associated with the ozone episode in the eastern United States.

[34] Some more input from the eastern United States was found by backward tracer calculations for the descending middle tropospheric layer during a period during the second half of 29 May (16:30 to 19:30 UTC, 5.5 to 7.5 km asl, see label 3 in Figure 1). The tracer plume, evaluated for 500 m above the surface, passed the east coast on 24 May, i.e., at the time when the second front ended the two-day 70-ppb period in New Jersey and Virginia. This front lifted the PBL air to about 5 km where it caught up with the main front because of rapid transport. This rapid airstream also contained some air from Texas and Mexico. The final height reached in the main WCB was 7 to 8 km.

[35] In order to complete the illustration of the full complexity of this period of transatlantic transport it should be mentioned that the major portion of the high-ozone air in the descending mid-tropospheric layer on 30 May (label 4 in Figure 1) had its principal North American sources in the Los-Angeles area and, again, Texas.

3.2. Case 2: 8–12 September 2000

[36] Figure 13 displays the results of the ozone soundings on 8–12 September 2000. The time series resembles that of case 1, which, together with other cases not presented here, seems to underline the reproducibility of the general pattern observed under developing anticyclonic conditions. Again, a stratospheric intrusion layer is seen (high ozone between 2 and 4 km (on 8 September), subtropical air with low ozone between 4 and roughly 7 km and North American air with high ozone above 7 km). However, the trajectory results (e.g., Figure 14, representing 9 September, 3:00 UTC) show that the ozone export from the United States does not proceed perpendicularly, but almost parallel to the North

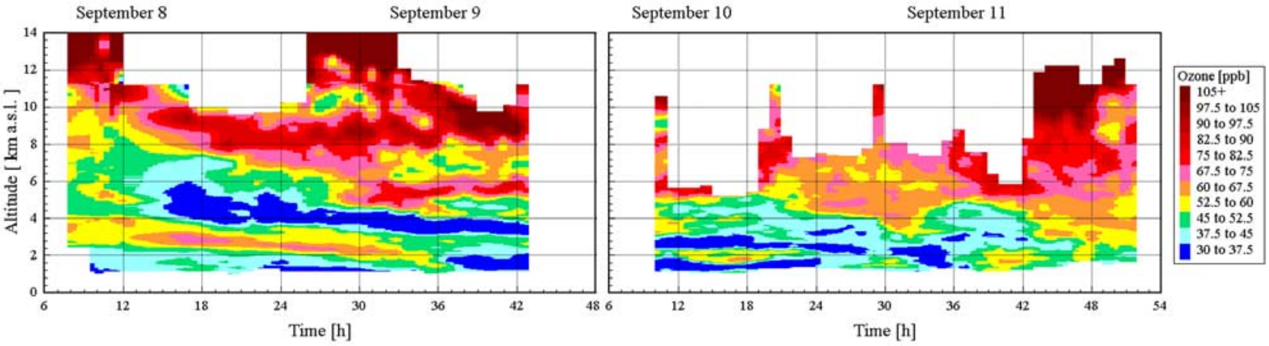

Figure 13. Lidar series 8–12 September 2000; the data gap between 9 and 10 September is due to interference by the IFU safety guard.

American east coast. The lowest elevations (below 4 km) are reached over Georgia and Alabama for arrival over Garmisch-Partenkirchen on 9 September, eight to ten days backward in time. The EPA maps for this area by the end of August show ozone mixing ratios exceeding 80 ppb, in part even reaching 100 ppb.

[37] The MOZAIC profiles for Frankfurt for 8 September show relative humidity values above 50%, sometimes even around 100%, in particular in the low-ozone layer from the subtropical Atlantic above the stratospheric intrusion. During the following days, similarly to case 1, increasingly dry layers are found in the middle troposphere, e.g., at about 6 km on 9 September, with some co-incidence with the elevated ozone measured with the lidar next to this height. As in case 1 a FLEXPART simulation with stratospheric tracer (resembling that in Figure 11) shows just minor stratospheric contributions from the North Atlantic.

[38] The export pattern is complicated by the presence of a sequence of fronts. As in case 1 there is no significant import from the central part of the eastern United States during the period of observation. The EPA data for the eastern United States show very high ozone values throughout much of August, too early with respect to our measurements. After 27 August, because of a frontal passage the values dropped to about 60 ppb. This is confirmed by MOZAIC profiles for the airports of Atlanta, New York and Boston. After 4 September the ozone concentrations in the eastern United States decreased even further because of another air mass change.

[39] A long WCB is formed over the Atlantic, which is nicely documented by the GOES-East images. Figure 15 shows an example from 6 September, 12:15 UTC, which is the beginning of the most important period for the observations above our site. The MOZAIC flights through the presumed outflow region of the WCB show high humidity and ozone mixing ratios of almost 100 ppb for the days before the lidar measurements. Behind the front an extremely large dry zone is seen, covering about half of the United States and ending in a long streamer toward Europe. However, the backward trajectories do not indicate a significant advection of air from this dry area to Garmisch-Partenkirchen. A similar (though less impressive) dry streamer had formed on 23 May 1996 (case 1).

[40] Figure 16 shows the result of a FLEXPART simulation of the North America NO_x tracer transport. As in the May 1996 case (case 1), most of the PBL air is concentrated in the upper (above 8.5 km) and middle troposphere (3 to 6 km) over IFU, roughly between noon of 8 September and noon of 11 September (afterward, some stratospheric influence was found by a separate North Atlantic stratospheric tracer calculation). The simulated layers nicely coincide with the high-ozone layers in the lidar time series, but the NO_x tracer contributions look moderate. However, in this case the trajectories (see Figure 14) show a mesoscale circulation over the North American east coast that may not be fully resolved by the ECMWF data. Indeed, the trajectories are likely not to reach the PBL, while in reality convection would have helped to lift up material from the surface. Although FLEXPART contains additional parameterizations to estimate the effects of subgrid-scale convection, the vertical transport may have been underestimated in this case. The GOES-East images, indeed, give evidence of enhanced convective activity in Alabama and its surroundings over several days in early September (see also Figure 15).

[41] As in case 1 some tracer from the North American PBL is found in the subtropical air layer. This contribution is rather significant on 8 September, before the beginning of the low-ozone period. The maximum coincides with an ozone peak in the lidar measurements.

[42] In contrast to the upper tropospheric ozone layers, which are mostly traced back to Alabama, the layer between 5 and 6 km on 9 September could not be identified by the trajectories, which are widely spread and become sparse on their way backward to the western United States. A backward tracer simulation for this layer was started between 7 and 11 UTC and identified most of the southern United States as a moderate source region. However, the majority of the tracers remained over the Atlantic throughout the calculation period ($\leq$32°S, between 0° and 65°W, back to 27 August). This indicates some overlap with the subtropical air mass for the start box selected and potentially some ozone input from further east.

[43] Similarly to case 1, significant amounts of North America tracer were found in the tracer results above 4 km before the observational period. This may be seen in Figure 16 for the first half of 7 September and also on the two days preceding the period shown in Figure 16. A backward tracer

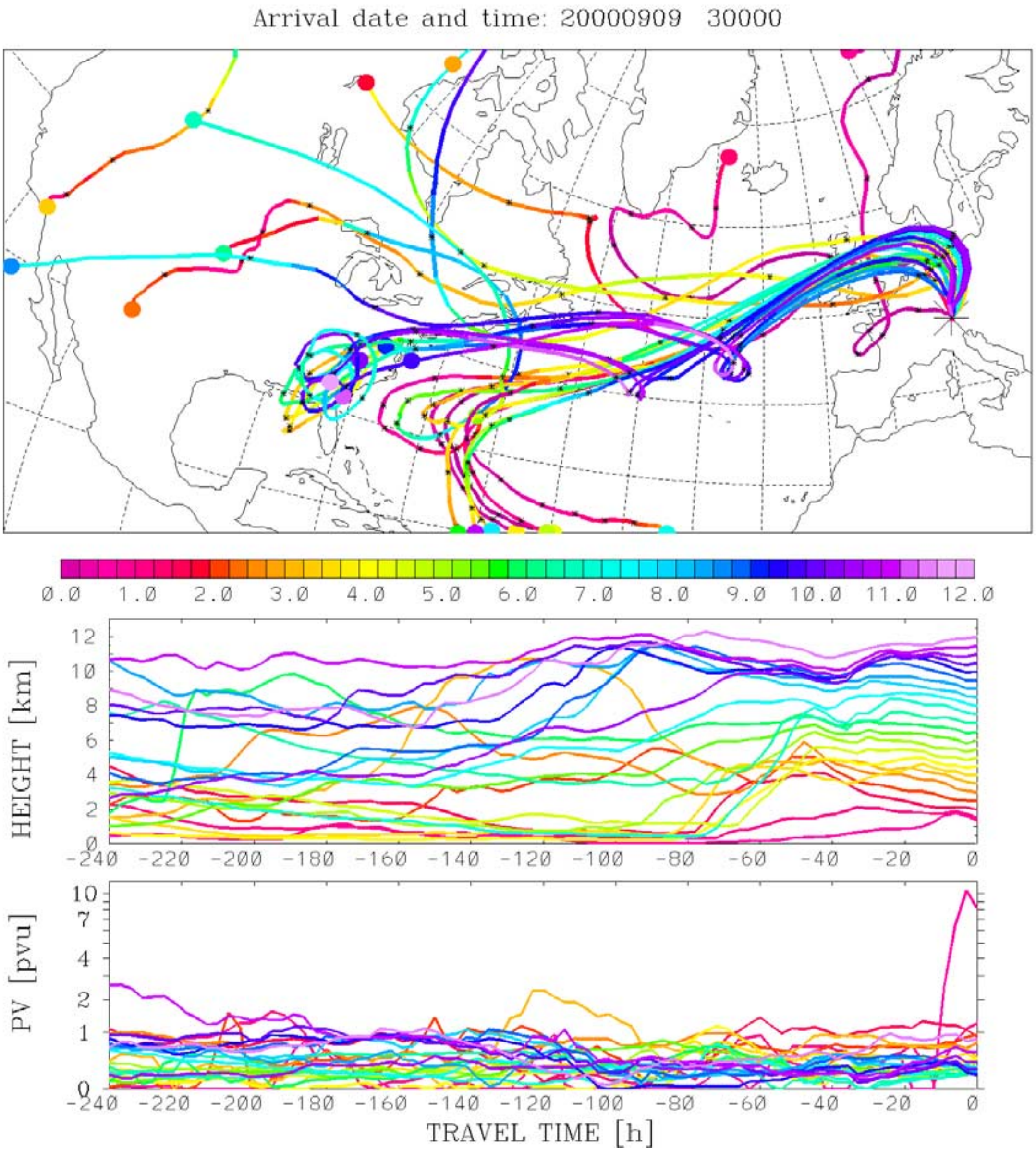

Figure 14. FLEXTRA trajectories for 9 September 2000, 3:00 UTC; for details, see Figure 2.

calculation for the late morning of 7 September shows input from the eastern United States three to twelve days backward in time, in some overlap with the ozone episode mentioned above.

3.3. Case 3: 26–30 May 1999

[44] The measurements discussed in the following were carried out as a part of the VOTALP "Munich" field campaign devoted to the advection of air pollution from the Munich urban area to the Alps. Measurements were made with both the stationary ozone and aerosol lidars. Because of the absence of a normal-strength valley wind following a weekend of disastrous local flooding there was no clear evidence of an urban plume streaming upward the Loisach valley and reaching Garmisch-Partenkirchen, perhaps with the exception of 27 May. Instead, the campaign yielded the highest ozone mixing ratios in the middle and upper free troposphere persisting over several days ever seen over Garmisch-Partenkirchen since lidar measurements have been carried out (Figure 17). The peak ozone mixing ratios were of the order of 130 ppb.

[45] Although, as seen in Figure 17, the measurements were interrupted several times (e.g., in the afternoon of 28 May because of rain), the general distribution of the high-ozone layers for the observational period (26–30 May) is rather well reproduced by a FLEXPART simulation with North America tracer (Figure 18) indicating a substantial contribution from the North American PBL.

[46] The backward trajectories show different advection pathways for the periods before and after 28 May, although the high-ozone layer above 6 km seems to be largely unchanged. The beginning of the observational period was quite different in its behavior from the other cases with long-range advection of layers with elevated ozone in the free troposphere (Figures 19 and 20 give examples for 26 May and 27 May, respectively). For this period the

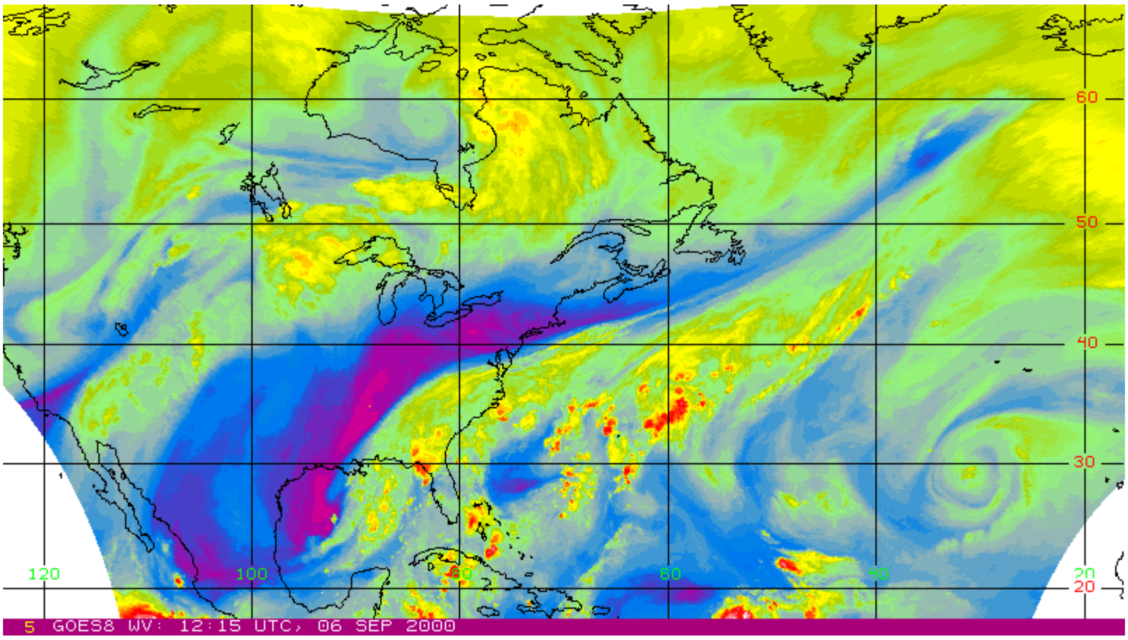

Figure 15. GOES-East water vapor image, 6 September, 12:15 UTC; for technical details, see section 2.4.

FLEXTRA trajectories show an almost straight principal advection pathway further north on 26 May. On 27 May the free-tropospheric ozone concentrations are particularly high, except for an "ozone hole" at about 5 km. The lower tropospheric mixing ratios were of the order of 70 ppb, which is quite remarkable for this time of the year. As we know from nearby ozone-sonde ascents (26–28 May, Paul-Scherrer-Institut, personal communication) the relative humidity below 4 km was of the order of 70% on 26 and 27 May. A significant European contribution to this layer is

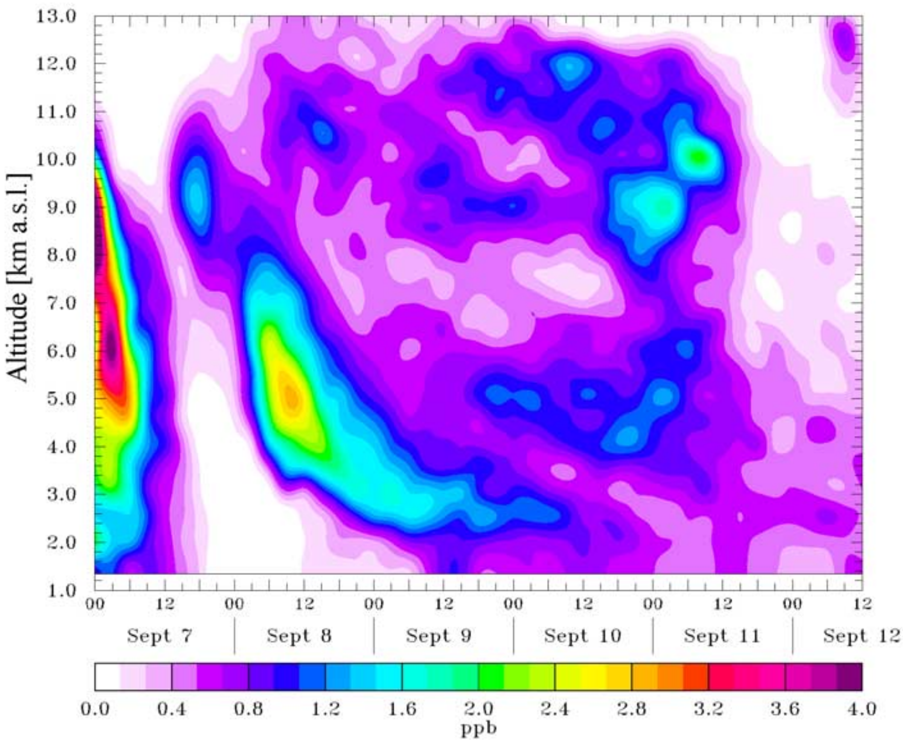

Figure 16. FLEXPART North America NO_x tracer run for the September 2000 case.

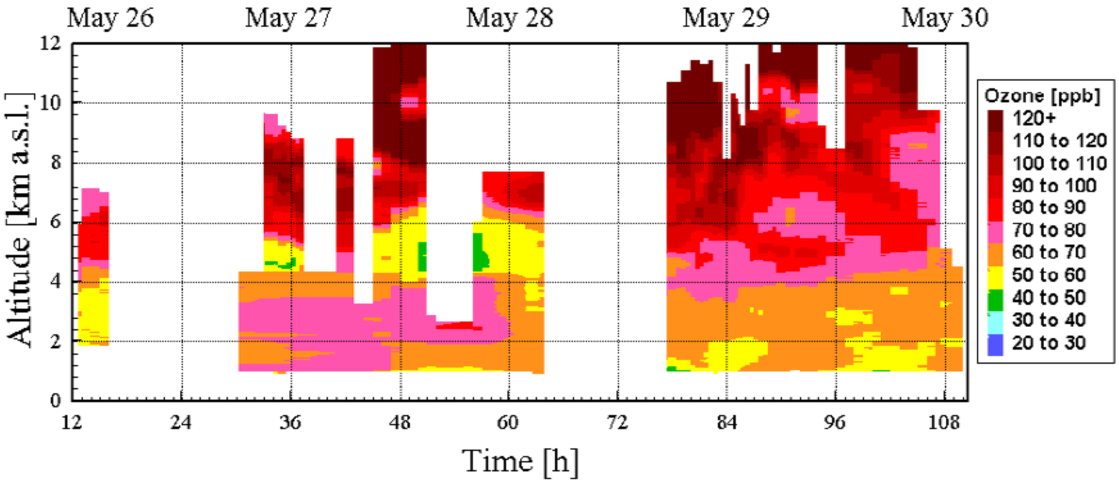

Figure 17. Ozone-lidar measurements 26–30 May 1999.

not likely, given a lower tropospheric air residence time over Europe of just one to two days. The trajectories arriving below 4 km roughly stay in this height range for ten days backward in time. The NO_y mixing ratios measured at Garmisch-Partenkirchen and the nearby Wank summit (1780 m asl) were mostly low with a moderate increase to 1–2 ppb on 27 May, which verifies the arrival of rather fresh marine air. For 27 May a lot of the lower tropospheric trajectories are distributed over the western part of the Atlantic or in the eastern United States toward ten days backward in time, the detailed pattern changing every few hours. A clear assignment is therefore difficult. The EPA maps show suitably high ozone mixing ratios only in the west of the Appalachian mountains, where some of the trajectories originate.

[47] The rather inhomogeneously structured layer above 5 to 6 km exhibited values between 80 and 130 ppb. Here, very low humidity (even below 15%) was registered with a clear anticorrelation with high ozone suggesting the presence of stratospheric or polluted upper tropospheric air. On the other hand, the high-ozone layers above 5 to 6 km were clearly correlated with aerosol throughout the operational period of the aerosol lidar (26–29 May), which indicates an origin of some of the air in the PBL. Figure 21 shows

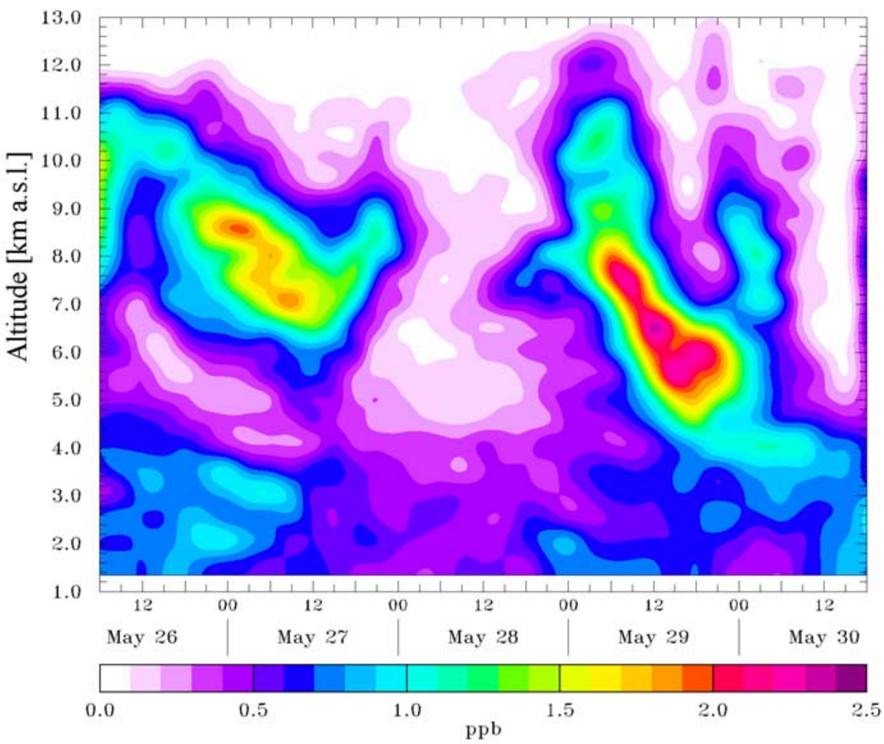

Figure 18. North America tracer simulation for 26–30 May 1999.

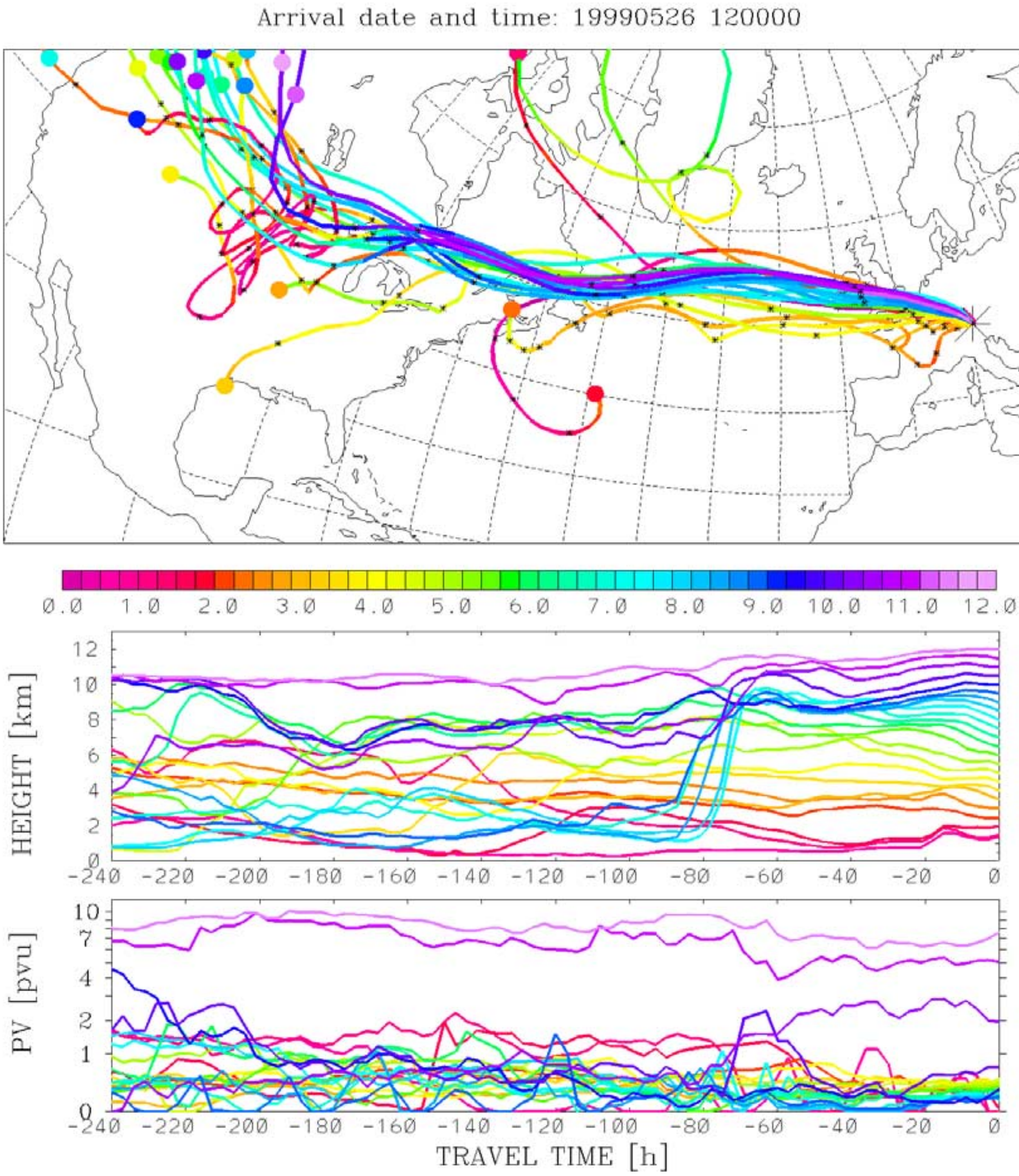

Figure 19. FLEXTRA backward trajectories for 26 May 1999, 12:00 UTC; for details, see Figure 2.

one example of a range-corrected backscatter signal (27 May, at about 12 CET) from the stratospheric aerosol lidar of IFU together with the almost simultaneously taken ozone profile from the ozone lidar. The agreement of the structures is obvious. However, a shift in height is seen with respect to the Paul-Scherrer sonde due to a growing distance of the sonde from the lidar. This discrepancy in layer position between sonde and lidar underlines the importance of side-by-side lidar profiling of relevant tracers (such as ozone, aerosol, water vapor) in a comparable air volume.

[48] The FLEXTRA trajectories (Figure 20, corresponding in time to the example in Figure 21) reveal a rather horizontal course of the trajectories below 4 km and above 8 km. In contrast, the trajectories between 4 and 8 km mostly originate over North America at altitudes below 2 km just four days backward in time. Just before the air mass arrival at the lidar site, the trajectories strongly descend, partly by more than 2 km, which could contribute to the low relative humidity in these layers (although the lowest humidity is seen between 7 and 9 km). Very interestingly, the export for the entire arrival period 26 May to 27 May took place in the same area to the south west of the Great Lakes by the beginning of 23 May (in UTC), as may be seen from the 24-h shift of the strong rise from the PBL in Figures 19 and 20.

[49] The situation in parts of the relevant area was derived from the limited number of EPA ozone maps available. On 22 May, ozone exceeded 60 ppb (mostly below 80 ppb in Kansas, Illinois and parts of Minnesota, with no indication of a pronounced rise when the front responsible for the air export moved across that region on its way to the east. The ozone concentrations significantly dropped after the frontal passage. It is obvious that the ozone mixing ratios below 80 ppb cannot account for the upper tropospheric maxima over Garmisch-Partenkirchen by the end of May. Therefore a pronounced entrainment of another component must be concluded. Indeed, some of the trajectories (see in particular

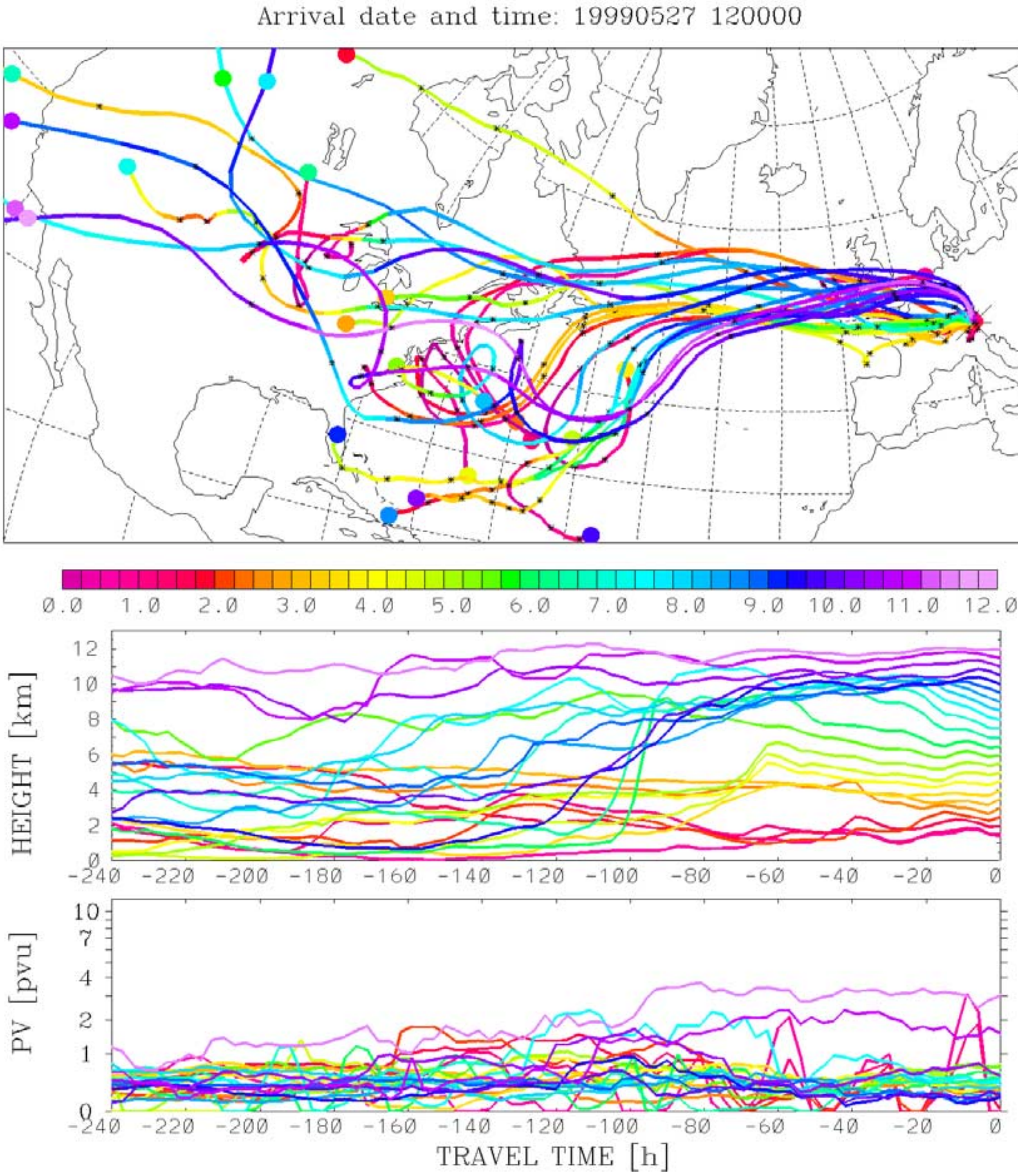

Figure 20. FLEXTRA backward trajectories for 27 May 1999, 12:00 UTC; for details, see Figure 2.

26 May, Figure 19) contributing to the high-ozone layer over Garmisch-Partenkirchen stay in the mid- to upper troposphere for ten days backward in time and may be traced back to the Pacific Ocean. As in case 1, the possibility of ozone import from East Asia or the stratosphere outside North America must be considered. The PV values along the trajectories are low, indicating no entrainment of fresh stratospheric air.

[50] Figure 22 shows the GOES-East water vapor image for 23 May, 0:15 UTC, showing the front to the south west of the Great Lakes lifting the PBL air to the upper troposphere reaching Garmisch-Partenkirchen IFU on 26 and 27 May. A very large convective cell formed during the following hours (more clearly visible in the 3:00-UTC image), which could have contributed some ozone because of chemical formation based on lightning-induced NO_x or by downward transport to the exported air layer from the stratosphere.

[51] The FLEXTRA trajectories for 28 May mostly originate over the Atlantic. For 29 May, many of the trajectories reach North America, but the trajectory pattern becomes more and more complex and it is difficult to identify a source region in the PBL. There is some indication that the western United States is contributing to the input, but the trajectories are widely spread and not long enough for a confirmation. At least the FLEXPART simulation (Figure 17) verifies the presence of significant amounts of North American PBL air on 29 May above 5 km. For 30 May, again, most of the trajectories come from the Atlantic Ocean.

4. Discussion and Conclusions

[52] The cases of intercontinental transport studied by *Stohl and Trickl* [1999] and in this paper have identified an important advection pathway for North American pollution

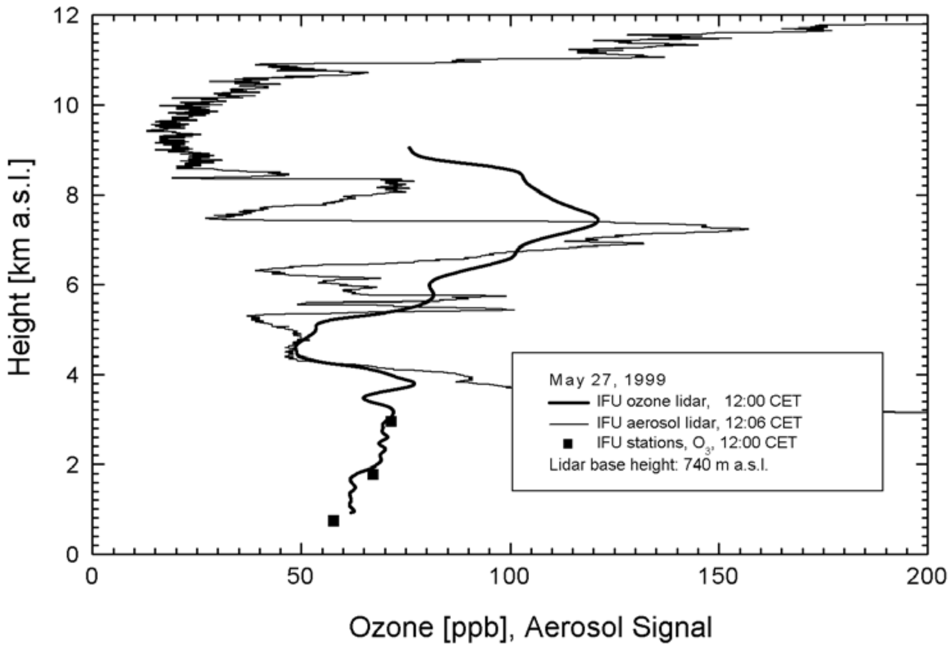

Figure 21. Results from ozone and aerosol lidar measurement on 27 May 1999, at about 12 CET (aerosol: range-corrected signal); moderate aerosol is seen in the free troposphere which correlates well with the ozone maxima. The pronounced backscatter signal above 11 km is caused by cirrus clouds.

to central Europe, resembling an S rotated by approximately 90°. In all cases of this type so far studied during the warm season a prefrontal high-ozone episode over the central part of the eastern United States with mixing ratios between 80 and 110 ppb was terminated by a cold-front passage within ten days prior to the observational period at Garmisch-Partenkirchen. The PBL air is lifted to up to more than 10 km with the WCB that originates in the polluted warm air ahead of the cold front. Rapid transport in the jet stream takes the WCB outflow toward Europe. Typically, the jet stream turns anticyclonically before reaching Europe, thus completing the rotated S. During observational periods starting with a deep stratospheric intrusion (May 1997, cases 1 and 2 and June 2001 [*Roelofs et al.*, 2003]) the normal atmospheric layering was found to be reversed over central Europe: Stratospheric air is found in the lower troposphere and initially lower tropospheric air arrives in the middle and upper troposphere.

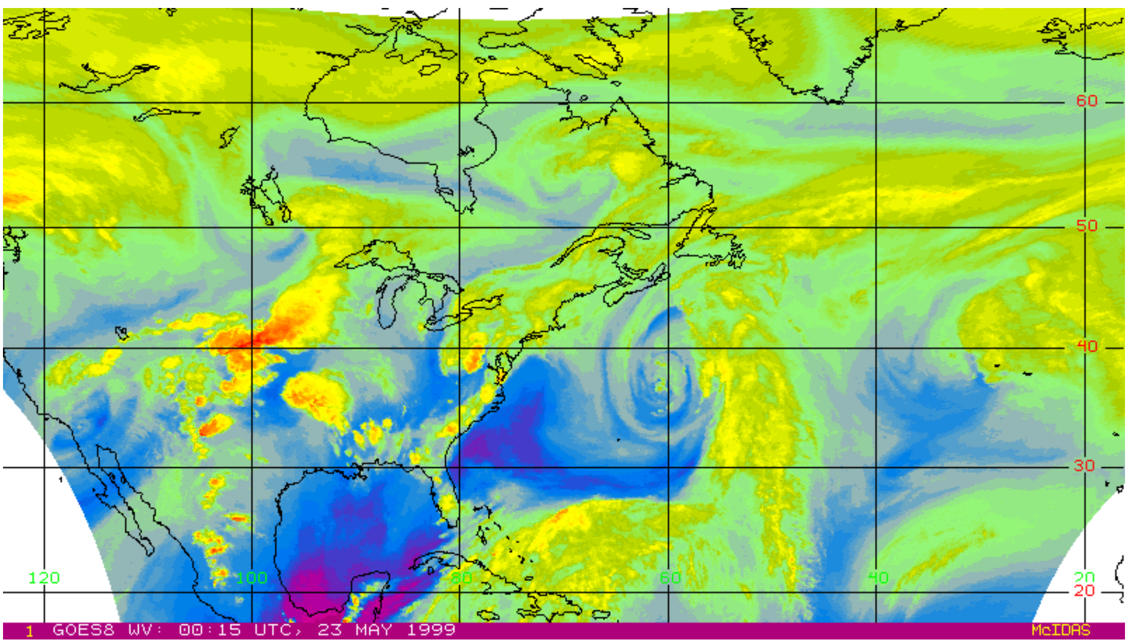

Figure 22. GOES-East image for 23 May 1999, 0:15 UTC; for technical details, see section 2.4.

[53] As shown by *Stohl et al.* [2002a] the outflow from the North American east coast, lifted in a WCB, mostly passes over central Europe at high altitudes. However, although the lidar observations at IFU presented in this paper show upper tropospheric ozone mixing ratios also in the 80 to 110 ppb range, the trajectory results reveal that, in contrast to the May 1997 case [*Stohl and Trickl*, 1999], the central part of the eastern United States is not always the principal source region during developing anticyclonic periods over central Europe. PBL contributions from other regions of the United States may be also important, and also very dry air masses entrained from beyond North America are found in these rapidly traveling high-ozone layers in the vicinity of the jet stream. In one case studied earlier, even rather low ozone import (50 to 60 ppb) to Europe was found due to advection from Florida [*Carnuth et al.*, 2002]. The tracer calculations reveal that the PBL contributions arriving in upper troposphere have a tendency of being concentrated in the early phase after the initial stratospheric air intrusion over central Europe. This might, in part, be ascribed to the strong coupling to the anticyclonically bent jet stream that transversely proceeds to Eastern Europe.

[54] The observational results and the tracer simulations indicate mixing of different contributions from the North American PBL air, some stratospheric air and, in part, ozone-rich air from beyond North America. As seen from the trajectory examples in this paper and also some of the satellite images, air masses of rather different origin are closely adjacent to each other during the final few days of travel to Europe. A close look at the trajectories shows that mixing mainly occurs in the vicinity of the fronts or the WCB outflow regions. Mixing of PBL and stratospheric air in the vicinity of fronts next to the North American east coast has been verified in several aircraft-based experiments [*Prados et al.*, 1999; *Parrish et al.*, 2000; *Cooper et al.*, 2001]. In addition, we have identified large thunderstorms in some of the satellite images, which could also contribute to the mixing of stratospheric and tropospheric air. The influence of lightning-produced NO_x to the build-up of ozone has been studied in numerous publications [e.g., *Liu et al.*, 1983; *Höller et al.*, 1999, and references therein]. *Jaegle et al.* [1998] report on efficient ozone production in the vicinity of the anvil of a mesoscale convective system over Wisconsin.

[55] Although the model simulations can suggest cases for which mixing has occurred, one cannot expect the model to be capable of simulating this mixing quantitatively. Mixing in the free troposphere is highly episodic, associated either with deep convection or with intermittent clear air turbulence (CAT). Although convective mixing is simulated, it is unlikely that it always occurs at the right location and time in the model. Similarly, free tropospheric turbulence is simulated continuously in the model (albeit it is stronger near the jet stream, because of the strong wind shear there), whereas in reality, extended periods without turbulence are interrupted by short bursts of strong turbulence. An improvement in this respect can be expected from ongoing work to implement a CAT scheme in FLEXPART. Such schemes are normally used for predicting CAT for air-traffic safety. However, with currently available meteorological data it will remain difficult to diagnose reliably turbulent bursts, or even to validate such a CAT scheme.

[56] The limited material so far available for periods during which a clear source-receptor relationship may be deduced suggests that there is no major change in ozone concentration during the passage across the ocean. This is a consequence of the fast transport that reduces the time for dilution [*Stohl*, 2001], but also indicates little chemical modification. Modeling studies by *Wild et al.* [1996] suggest that there might be some chemical transformation of the air mass on its way across the ocean. The results for the 300 hPa pressure level show an ozone increase by 20 to 30%, whereas those for 900 hPa do not exhibit substantial changes in the ozone concentration, with a small tendency for slow destruction. It is obviously important to consider the influence of removal processes in the WCB on the chemical composition of the air mass in order to understand the behavior in the subsequent phase in the upper troposphere. Indeed, NO_y, which is mostly in the form of HNO_3 in the lower troposphere, may be removed to a large extent during export from the PBL [*Cooper et al.*, 2001; *Stohl et al.*, 2002b], limiting the ozone-forming capacity in the upper troposphere. On the other hand, lightning, which can occur in a WCB, would enhance NO_x concentrations and, together with the hydrocarbons from surface sources, could result in efficient ozone production.

[57] Following the start of several of the episodes extremely dry air was observed above 5 km. The considerable thickness of these dry layers prevents them from being mixed with surrounding air. The maximum ozone values mostly exceed 110 ppb, suggesting that aged stratospheric air may have entered the troposphere far to the west (possibly even over the Pacific Ocean). The air travels at high speed and is, thus likely to be a part of the jet stream. The large layer width and the high ozone values differ from our observations in the lower troposphere where just 60 to 90 ppb are seen in the vertically confined stratospheric ozone tongues. The reduction in concentration in the case of the deep intrusions is mainly caused by horizontal spreading of the air tongues. It is important to note that during almost all short or extended direct intrusion periods we have found at least one strongly descending component reaching the Zugspitze summit [*Eisele et al.*, 1999]. If stratospheric air is entrained to these wide upper tropospheric airstreams the transfer mechanism must be rather special.

[58] An interpretation of the high ozone values above 5 km by inflow of stratospheric air is also somewhat impeded by the low PV values calculated by the trajectory model, although PV is obviously not an important quantity considering the potentially long transport in the troposphere speculated on. From a similar observation recently reported on by K. S. Law et al. (Evidence for anthropogenic influence over the central North Atlantic, *IGACtivities,* newsletter 24, pp. 17–19, 2001) the authors conclude that because of low PV and high NO_x and CN concentrations in their case, an origin of these air masses in the stratosphere is not obvious. We plan to extend our model simulations to include emissions from East Asia. These emissions are frequently transported to the upper troposphere over the northern Pacific in WCBs, which is the region some of the fast airstreams rich in ozone identified in the present study came from. Because of the extremely long distances these simulations are rather ambitious.

[59] Case 3 demonstrates that North American ozone can reach the free troposphere in various ways. A full interpretation of this case has not been possible, although some conclusions are drawn from the humidity and aerosol data. The data suggest a co-existence of PBL and stratospheric/upper tropospheric air in the same high-ozone air mass. However, source regions can only partly be identified and the way in which the stratospheric or polluted upper tropospheric air was mixed into the layer is not entirely obvious. Both an influence of long-range transport from Asia and of large-scale thunderstorms must be considered.

[60] Aerosol, as used for the interpretation of case 3, is a reasonable tracer for PBL air. However, the application of the aerosol backscatter coefficient does not yield a quantitative transport budget since aerosol may be subject to washout in the fronts. In fact, free-tropospheric aerosol measurements at 532 nm with the sensitive stratospheric aerosol lidar of IFU rarely yield aerosol structures exceeding the Rayleigh background by more than 20%. Because of the much lower sensitivity for aerosols at 313 nm caused by the much larger Rayleigh background, none of the backscatter profiles of the ozone lidar has yielded any clear evidence of aerosol in the layers of North American origin. Nevertheless, aerosol in the free troposphere indicates the presence of layers fed by long-range transport from a remote PBL. In a recent study within the German Aerosol Lidar Network three years of data from the stratospheric aerosol lidar of IFU provided by H. Jäger were analyzed [*Trickl and Wandinger*, 2001]. It was found that distinguishable aerosol signatures in the free troposphere (i.e., exceeding the Rayleigh contribution by at least 5%) occurred in about 40% of the measurements. A pronounced spring maximum was observed (65% of the cases between 21 March and 20 June). This stimulates an intriguing question about the role of long-range transport for the well-known spring maximum of ozone [*Monks*, 2000, and references therein].

[61] The observation of transatlantic transport of aerosol is, in contrast to that of ozone, nothing particularly new. In particular, IFU researchers reported on observations of aerosol from a dust outbreak in Colorado in February 1977 on the nearby summit Wank (1780 m asl), nicely verified by a satellite image [*Reiter et al.*, 1984]. Of course, the observation of aerosol does not necessarily mean input from the North American PBL. Also aerosol exported to the Atlantic Ocean from Africa may reach Europe after circulating around a high-pressure system [e.g., *Kreipl et al.*, 2001; *Carnuth et al.*, 2002].

[62] The observation of dust-loaded African air masses after export to the Atlantic Ocean is a rather rare event. In contrast, the advection of moist air with low ozone concentration from the subtropical Atlantic under the "S-shape" conditions is highly reproducible. Our first observation of this kind was made in 1991 [*Carnuth et al.*, 2002]. The results obtained during VOTALP and more recent measurements have repeatedly shown this component in the WCB air mass, with mixing ratios between 25 and 40 ppb. Ozone values as low as 18 ppb in tropical air transported in WCBs from the Pacific northward to the United States were reported by *Grant et al.* [2000].

[63] The wide range of relative humidity values of tropospheric and stratospheric origin in the different layers makes water vapor an important tracer for transport studies. As a consequence, we plan to carry out simultaneous lidar measurements of ozone, aerosol and water vapor in forthcoming studies of long-range transport within the German research program Atmosphärenforschung 2000. Furthermore, with the additional benefit of being able to include model forecast data on a daily basis for up to 72 h after the respective verification times, the investigations will become more systematic, which allows us to estimate the relative importance of the different advection pathways. These studies will also focus on the importance of large-scale North American forest fires on the extent of air pollution over central Europe [e.g., *Wofsy et al.*, 1994; *Wotawa and Trainer*, 2000; *Forster et al.*, 2001; *Wotawa et al.*, 2001].

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