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Impact of transatlantic transport episodes on summertime ozone in Europe

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Abstract

This paper reports on the transport of ozone (O_3) and related species over the North Atlantic ocean and its impact on Europe. Measurements of nitrogen dioxide (NO_2) and carbon monoxide (CO) columns from the GOME and MOPITT satellite instruments, respectively, are used in conjunction with the GEOS-CHEM global model of transport and tropospheric chemistry to identify the major events of long range transport that reach Europe over the course of summer 2000. Sensitivity model simulations are used to analyse observed O_3 distributions with respect to the impact of long range transport events. For that purpose, we used in-situ O_3 observations taken at the mountain site of Jungfraujoch as well as O_3 vertical profiles taken in the vicinity of central European cities. Over the course of summer 2000, we identified 9 major episodes of transatlantic pollution transport; 7 events are associated with transient cyclones while 2 events occur through zonal transport (e.g. by advection in the strong low-level westerly winds established in summer between the Azores anticyclone and transient cyclones). We find that on average three episodes occur per month with the strongest ones being in June. The number and frequency of long range transport events that reach Europe are driven by the position and strength of the Azores anticyclone. Model sensitivity simulations indicate that the summer mean North American O_3 contribution ranges from 3 to 5 ppb (7-11%) in the planetary boundary layer and 10 to 13 ppb (18-23%) in the middle and upper troposphere. During particular episodes, North American sources can result in O_3 enhancements up to 25-28 ppb in the layer between 800-600 hPa and 10-12 ppb in the boundary layer. The impact of the zonal transport events on O_3 distribution over Europe is more clearly seen below 700 hPa as they tend to transport pollution at lower levels while the events associated with transient cyclones are more likely to have an impact on the middle and upper troposphere (i.e. above 600 hPa). The air mass origins found in the GEOS-CHEM model are clearly confirmed by back trajectory analyses. During most of the 9 events, a strong contribution in North American O_3 is in general associated with only little European O_3 and vice-versa (in particular at the Jungfraujoch). A substantial North American contribution (e.g., 30% or higher) to O_3 over Europe does not always result in pronounced O_3 enhancements in the observations during our period of study.

Keywords

europe, ozone, episodes, impact, transatlantic, summertime, transport

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Impact of transatlantic transport episodes on summertime ozone in Europe

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1 Introduction

Long range transport (LRT) of ozone (O_3) and related species in the Northern Hemisphere is of major importance since the main anthropogenic sources are the populated regions of North America, Europe and Asia. In particular, transatlantic transport of O_3 is raising a lot of interest because of the relatively short distance between North America and Europe (Wild and Akimoto, 2001).

Pollution export from North America is driven by mid-latitudes cyclones (Stohl, 2001). The Warm Conveyor Belt (WCB), one of the four airstreams of the mid-latitude cyclones, is the most important pathway for rapid and direct intercontinental transport (Cooper et al., 2004) as reported in several case studies (e.g. Wild et al., 1996; Stohl and Trickl, 1999; Cooper et al., 2001; Trickl et al., 2003; Stohl et al., 2003a; Huntrieser et al., 2005). Deep convection is also an efficient pathway to ventilate the boundary layer (Li et al., 2005; Auvray and Bey, 2005), especially over

the south-eastern and central U.S. in summer. Pollution export occurs predominantly in the latitude range between 30 and 55° N, with little outflow poleward of 60° N (Li et al., 2005). The pollution pathways over the North Atlantic depend strongly on the position and strength of the Azores anticyclone (Li et al., 2002). Stohl (2004) found that the pollution transported in summertime in the middle and upper troposphere reaches Europe north of 55° N while the pollution transported in the lower troposphere ends up at about 50° N. The temporal scale of LRT varies between 1 and 10 days (Stohl et al., 2003b; Wild and Akimoto, 2001) with important seasonal variation. The transport is much faster in winter compared to summer, but import of O_3 into Europe is at a maximum in spring and summer, reflecting the seasonal variation in photochemical activity and in export pathways (Auvray and Bey, 2005).

A quantitative understanding of the contribution of the O_3 transported from upwind regions versus that produced locally is becoming necessary because LRT can have a substantial impact on air quality (e.g. Li et al., 2002; Hudman et al., 2004) and on the effect of O_3 precursor emission reduction scenarios (e.g. Jacob et al., 1999; Langmann et al., 2003). For example, previous modelling studies indicated that North American emissions contribute by 11% to the O_3 annual average burden over Europe while European sources only contribute by 9% (Auvray and Bey, 2005). Li et al. (2002) suggested that 20% of the violations of the European O_3 standard would not have occurred in absence of anthropogenic emissions from North America. The main objective of the present paper is to quantify the impact of LRT events on the O_3 distribution over Europe in summer using various observational dataset as well as modelling tools. We focus on summer as it is the season of interest for the air quality issues.

Li et al. (2005) reported 21 export events out of North America (with a frequency of every 4 to 5 days) over the course of the summer 2000. We first expand on this later work to identify the major events of LRT that entered Europe during the summer 2000. To achieve this, we use satellite retrievals of nitrogen dioxide (NO_2) and carbon monoxide (CO) and simulations performed with the global chemical transport model GEOS-CHEM. The MOPITT CO observations provide a good signature of continental outflow, while the GOME NO_2 observations provide an additional indication of the chemical environment in the plumes. We then examine O_3 concentrations at the mountain site Jungfraujoch (JFJ, 3580 m a.s.l., Switzerland) as well as O_3 vertical profiles provided by the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC) program over several European cities with the objective of identifying and quantifying the signal associated with the LRT events. We used the JFJ observations as daily O_3 concentrations are available on this site; in addition, as the JFJ site is located in the free troposphere (FT) of central Europe, it is potentially a receptor point for most of the LRT events entering Europe. The MOZAIC profiles allow us to examine the impact of LRT events throughout the troposphere with good temporal and spatial resolution.

This paper is organized as follows. Section 2 presents the different datasets used, i.e. satellite retrievals of CO and NO_2 from the MOPITT and GOME instruments respectively, the ground based observations at JFJ, and the MOZAIC vertical profiles. The GEOS-CHEM model used in this work is described in Sect. 3. The LRT episodes entering Europe in summer 2000 are described in Sect. 4. The impact of North American plumes on O_3 distributions in Europe is examined in Sect. 5. Conclusions are given in Sect. 6.

2 Observation datasets

2.1 Tropospheric column of carbon monoxide from MOPITT

The MOPITT instrument flies on the NASA EOS Terra satellite which has a near-polar, sun-synchronous orbit with an inclination of 98.2° with respect to the equator, an altitude of 705 km, a descending node crossing time at 10:30 LT and a global coverage in approximately three days. MOPITT is a nadir sounding instrument with field-of-view $22\times22\,km^2$ (http://www.atmosp.physics.utoronto.ca/MOPITT/mdd_93/index.html). A detailed description of the MOPITT CO retrieval algorithm can be found in Deeter et al. (2003). The MOPITT Version 3 data is used for this study. In this paper, the model CO tropospheric column (integrated from the surface to the model tropopause) is only compared qualitatively to the MOPITT column.

2.2 Tropospheric column of nitrogen dioxide from GOME

The GOME instrument flies on European Space Agency ERS-2 satellite, which has a sun-synchronous polar orbit with an inclination of 98°, a mean altitude of 780 km, equator crossing at 10:30 LT and global coverage of 3 days. GOME has a nadir-scanning ultraviolet and visible spectrometer for global monitoring of total atmospheric ozone and nitrogen dioxide (Burrows et al., 1999). The instrument has a field-of-view of $320\times40\,km^2$ ($2.8°\times0.14°$). The version 2 retrieval of tropospheric NO_2 columns from GOME used here is based on the version 1 algorithm described in Martin et al. (2002). The typical uncertainty estimate for each observation is $\pm(1\times10^{15}\,molec\,cm^{-2}+40\%)$. After accounting for random errors, the monthly mean uncertainty reduces to $\pm(5\times10^{14}\,molec\,cm^{-2}+30\%)$. As for the CO column, the model NO_2 column is integrated from the surface to the tropopause.

2.3 Surface observations of O_3 at the Jungfraujoch

O_3 measurements at JFJ are performed with a UV absorption instrument. Estimated standard uncertainty for hourly

averages is 1.2 ppb for values below 60 ppb and 2% for values greater than 60 ppb. The JFJ site is at an altitude of 3580 m a.s.l. (650 hPa) and is surrounded by industrialized regions. The JFJ is situated in the free troposphere 57% of the time according to Li et al. (2006)[1], and is influenced by planetary boundary layer air (Baltensperger et al., 1997; Zellweger et al., 2002) especially during the warmer months. The wind direction at the site is influenced by the northeast-southwest orientation of the local alpine watershed, resulting in an average annual wind frequency of 60% from north-west and 30% from south-east (Baltensperger et al., 1997).

2.4 O_3 vertical profiles from MOZAIC

MOZAIC O_3 concentrations are measured on board of commercial aircraft by a dual beam UV absorption instrument. The overall precision of the data is $\pm$(2 ppb+2%). O_3 measurements have been validated using ozonesonde data. In the free troposphere (800–300 hPa) the agreement found (3 to 13%) was well within the uncertainty of the two techniques (Thouret et al., 1998). In this manuscript we use MOZAIC profiles over European (e.g. Paris) airports which are available on a daily basis.

3 Model description

In this manuscript, we used the GEOS-CHEM model, version 5-07-08 with an horizontal resolution of 2° of latitude by 2.5° of longitude and 30 vertical levels from the surface to 0.01 hPa, including 18 levels in the troposphere (http://www-as.harvard.edu/chemistry/trop/geos/index.html). A brief description of the model is given below. Further details can be found in Bey et al. (2001), Martin et al. (2003) and Park et al. (2004).

The model is driven by meteorological fields provided by the Global Circulation Model GEOS-3 of the Global Modeling and Assimilation Office (GMAO) at NASA with a temporal resolution of 3 and 6 h. 31 tracers are transported to provide a comprehensive description of the NO_x-O_x-hydrocarbon chemistry and nitrate-ammonium-sulphate aerosol chemistry. A total of 300 chemical reactions are taken into account in the chemical mechanism for about 80 different species, as described in Fiore et al. (2003). The photolysis rates are calculated with the Fast-J algorithm (Wild et al., 2000), which accounts for clouds and aerosols. When not provided by the model itself, aerosols are taken from the GOCART model (Chin et al., 2002) and coupled to GEOS-CHEM model as described in Martin et al. (2003). Dry deposition velocities are computed using a resistance-in-series model described in Wesely (1989) and modified by Wang et al. (1998). Wet deposition is applied to aerosols, HNO_3 and H_2O_2 as described in Liu et al. (2001). Anthropogenic emissions of NO_x, CO and hydrocarbons are prescribed as monthly mean fluxes. Natural emissions from plants and NO_x sources from lightning are calculated online with different parameterizations in order to capture their temporal and spatial variability. Biomass burning emissions are included on monthly bases as described in Duncan et al. (2003).

We present here the results from a standard simulation for 2000 obtained after a one-year spin-up. Two sensitivity simulations were also conducted with anthropogenic emissions of O_3 precursors from North America and from Europe turned off. Subtraction of these results from those of the standard simulation allow us to quantify the fraction of O_3 due to North American and European anthropogenic emissions (further referred to as North American and European O_3, respectivelly). In addition, we performed a "tagged" O_3 simulation to separate the stratospheric contribution. In that simulation, the production and loss rates archived from the standard simulation are used to drive an off-line simulation in which different "tagged" O_3 tracers are transported according to their region of production.

The model has been previously the subject of several global evaluations (e.g., Bey et al., 2001; Martin et al., 2003). Regional validations were presented by Fiore et al. (2002) and Li et al. (2005) for the United States, Li et al. (2002) for the Western North Atlantic Ocean, and Duncan and Bey (2004) and Auvray and Bey (2005) for Europe. Evaluations of O_3 and CO on a global scale do not show obvious biases. However, monthly mean O_3 concentrations show too little seasonal variation especially at the 500 hPa level (Bey et al., 2001; Fusco and Logan, 2003). Over Europe, monthly mean CO concentrations at the surface were found to be too low in winter and spring by 20 ppb and too high in summer and fall by 20 ppb (Auvray and Bey, 2005). Auvray and Bey (2005) showed that the GEOS-CHEM model captures well the major features of enhanced O_3 concentrations associated with two LRT events of North American origin observed over the eastern Atlantic Ocean and in the FT over Europe. Further evaluation of the model is presented in Sect. 5 by comparison to O_3 observed at JFJ and O_3 MOZAIC profiles. Note that in this paper, we do not present a quantitative evaluation of the model against MOPITT and GOME observations but we use these observations to examine the occurrence of LRT episodes in a qualitative way. Comparison of simulated CO columns to MOPITT and simulated NO_2 columns to GOME are discussed elsewhere (Li et al., 2005; Martin et al., 2003).

4 LRT episodes over the North Atlantic in summer 2000

In summer the airflow circulation over the North Atlantic ocean is driven by the persistent Azores anticyclone and its

[1] Li, Y., Staehelin, J., Auvray, M., Bey, I., and Schultz, M.: Comparison between numerical simulations of two 3-D global models (GEOS-CHEM and MOZART) with ozone observations at Jungfraujoch (Switzerland) and ozone sondes from Payerne, Atmos. Environ., submitted, 2006.

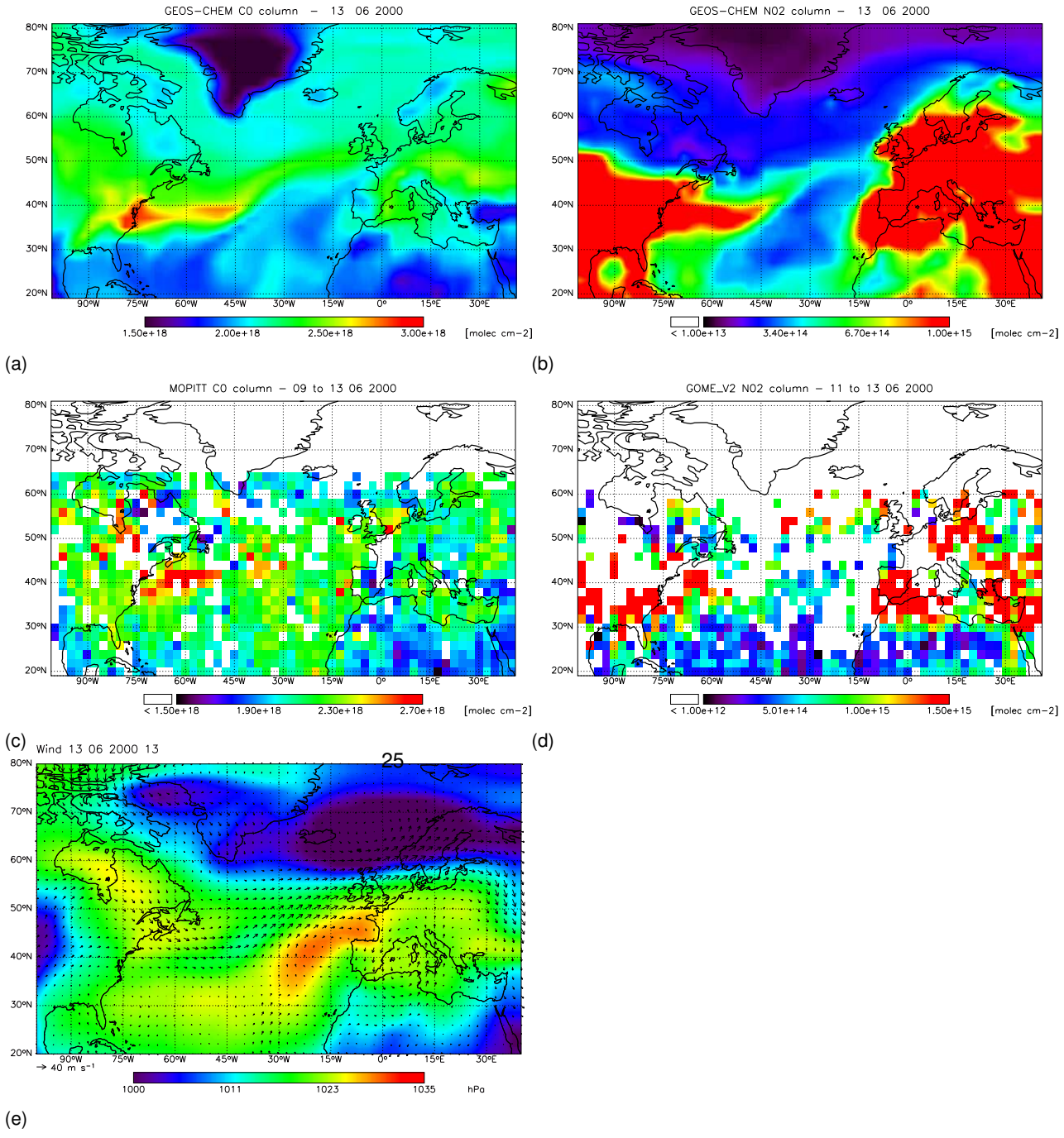

Fig. 1. Illustration of LRT1 event: **(a)** GEOS-CHEM CO column on 13 June, **(b)** GEOS-CHEM NO_2 column on 13 June, **(c)** MOPITT CO column on 9–13 June, **(d)** GOME NO_2 column on 11–13 June and **(e)** GEOS-CHEM mean sea level pressure and 500 hPa winds on 13 June.

position and strength control the transatlantic transport. In June 2000 the Azores anticyclone is centered at around 30° N and a strong westerly zonal flow is established between 45°–55° N. In July the Azores anticyclone expands to the north (50° N at 850 hPa) covering completely the North Atlantic and a strong south-west flow dominates along the east coast of the North America. In August the anticyclone retreats south of 40° N and a westerly zonal flow is predominant at 500 hPa between 40°–50° N.

Li et al. (2005) reported 21 export events out off North America into the North Atlantic over the course of the months of June, July and August 2000. We build on their

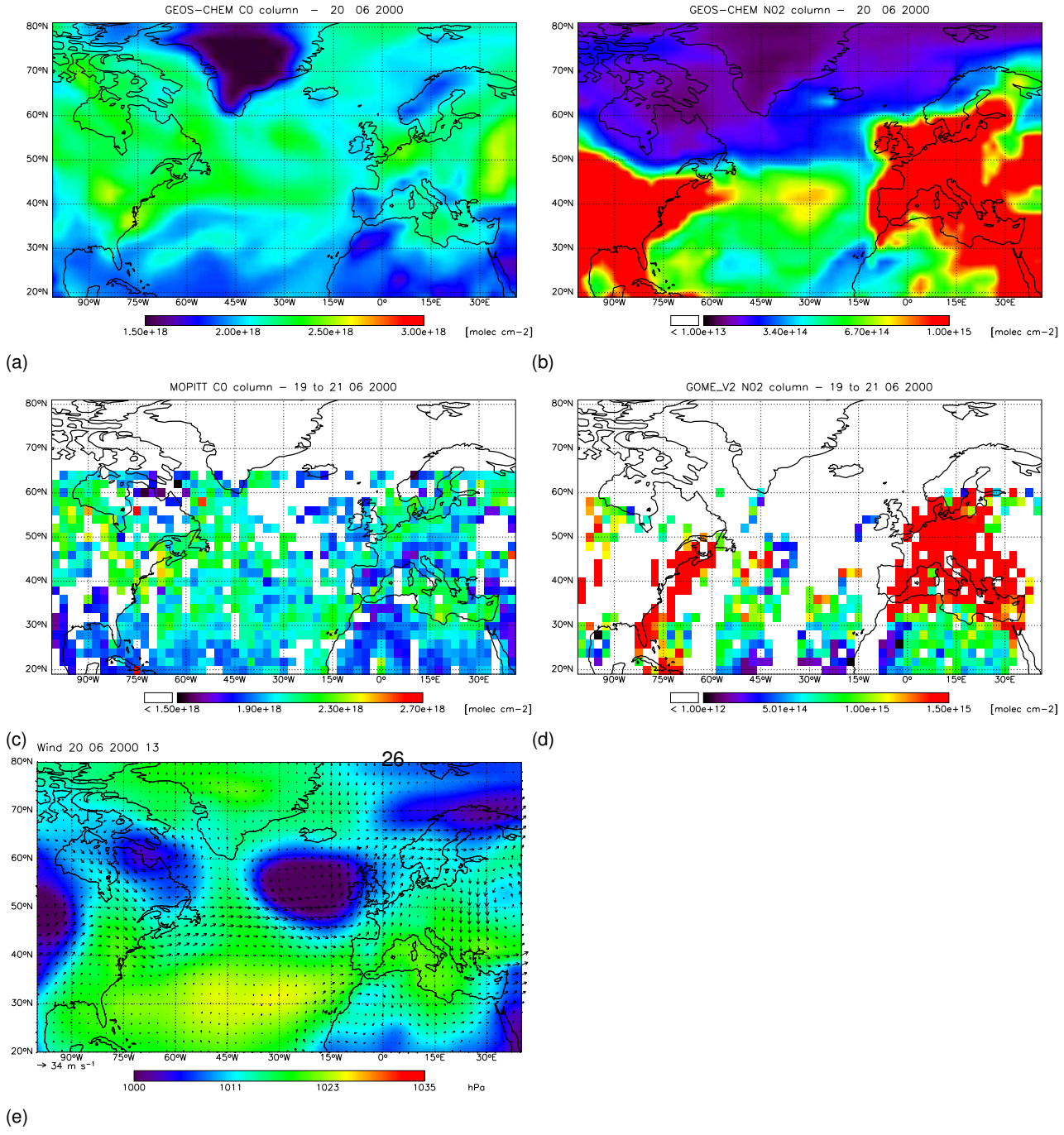

Fig. 2. Illustration of LRT2 event: **(a)** GEOS-CHEM CO column on 20 June, **(b)** GEOS-CHEM NO_2 column on 20 June, **(c)** MOPITT CO column on 19–21 June, **(d)** GOME NO_2 column on 19–21 June and **(e)** GEOS-CHEM mean sea level pressure and 500 hPa winds on 20 June.

work and examined daily maps of CO and NO_2 from MOPITT and GOME, respectively, and from GEOS-CHEM over the North Atlantic to identify the LRT events that actually reach Europe. After leaving North America, the plumes can either i) travel in the North American cyclones tracking poleward and thus reach Europe at high latitudes; ii) be transported zonally between 40° and 55° N directly to Europe; iii) be incorporated into the Azores anticyclone and reach Europe at mid-latitudes. However, not all the plumes that leave North America reach Europe. In some cases, for example if the plumes are transported at the periphery of the Azores high, they may travel back toward North America and never

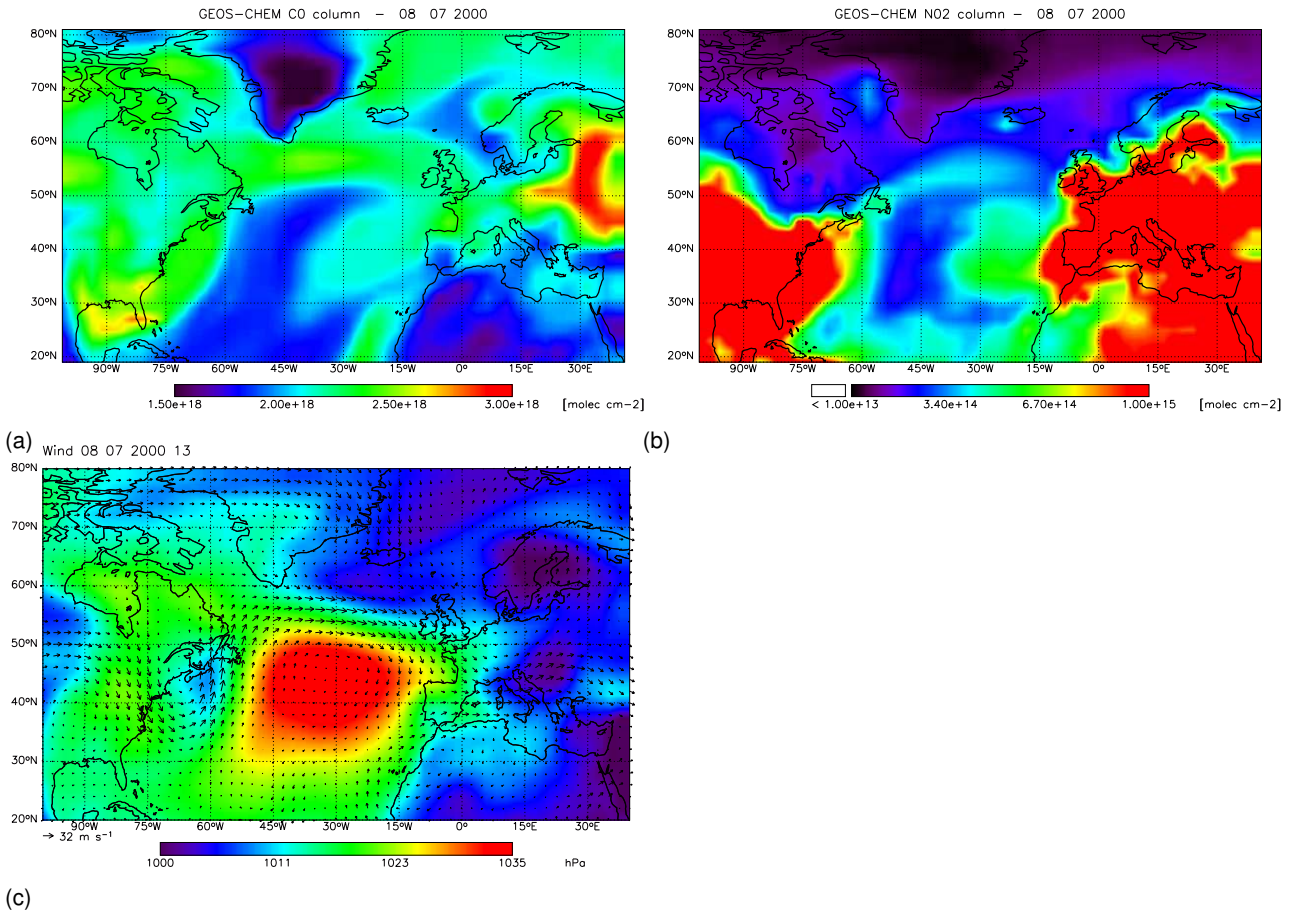

Fig. 3. Illustration of LRT4 event: **(a)** GEOS-CHEM CO column on 8 July, **(b)** GEOS-CHEM NO_2 column on 8 July, **(c)** GEOS-CHEM mean sea level pressure and 500 hPa winds on 8 July.

reach Europe. We identify a total of 9 major LRT episodes over the summer 2000 that reach central Europe. Table 1 summarises the places and dates at which the plumes travel and enter Europe. We also provide a qualitative characteristic of the plume, i.e. anthropogenic and/or biomass burning, when ATSR fire counts are visible in the vicinity of the starting place. Seven of the LRT episodes are associated with transport in a cyclone. Two episodes are associated with a very rapid zonal transport similar to what is described in Stohl et al. (2003b). Those episodes are only partly confirmed in the MOPITT and GOME data due to lack of observations in the cloudy regions of the cyclone. We find that on average three episodes occur per month with the strongest one being in June 2000. Four representative episodes are described in the following to illustrate the typical pathways.

The first episode of transatlantic LRT, denoted as LRT1, is presented in Fig. 1. On 8–9 June, the Azores anticyclone develops and cyclogenesis takes place in the Hudson Bay. On 10–13 June, the anticyclone extends toward southern Europe, while the low pressure system moves northeastward toward Greenland. Strong west-southwest winds (20 m/s at 850 hPa and 40 m/s at 500 hPa) develop at 35° to 50° N between the high and low systems. Through this "express highway" (Stohl et al., 2003b) the anthropogenic emissions from U.S. are directed toward UK and Scandinavia (Fig. 1e). LRT1 is clearly seen in the MOPITT and GOME observations at 40° N (Fig. 1c and Fig. 1d). NO_2 observations (Fig. 1d) also show an enhancement at higher latitudes (between 50° and 60° N) which is not seen in the model. This enhancement does not appear to be associated with transport event and may be due to lightning production in the cyclone.

The second episode (LRT2) occurred in the period of 16–21 June (Fig. 2). In the model CO plume is transported by an Atlantic cyclone that is moving to the north east (above 50° N). While reaching Iceland, the cyclone deepens and moves toward Europe reaching the UK, France and Portugal on 21 June (Fig. 2e) and continuing toward Norway where it occludes. The CO enhancement could not be confirmed in the MOPITT retrieval (Fig. 2c) due to observation gaps. Note that a NO_2 enhancement is seen in both the observations and the model at about 42° N. We found that this enhancement is restricted to the lower troposphere and is likely

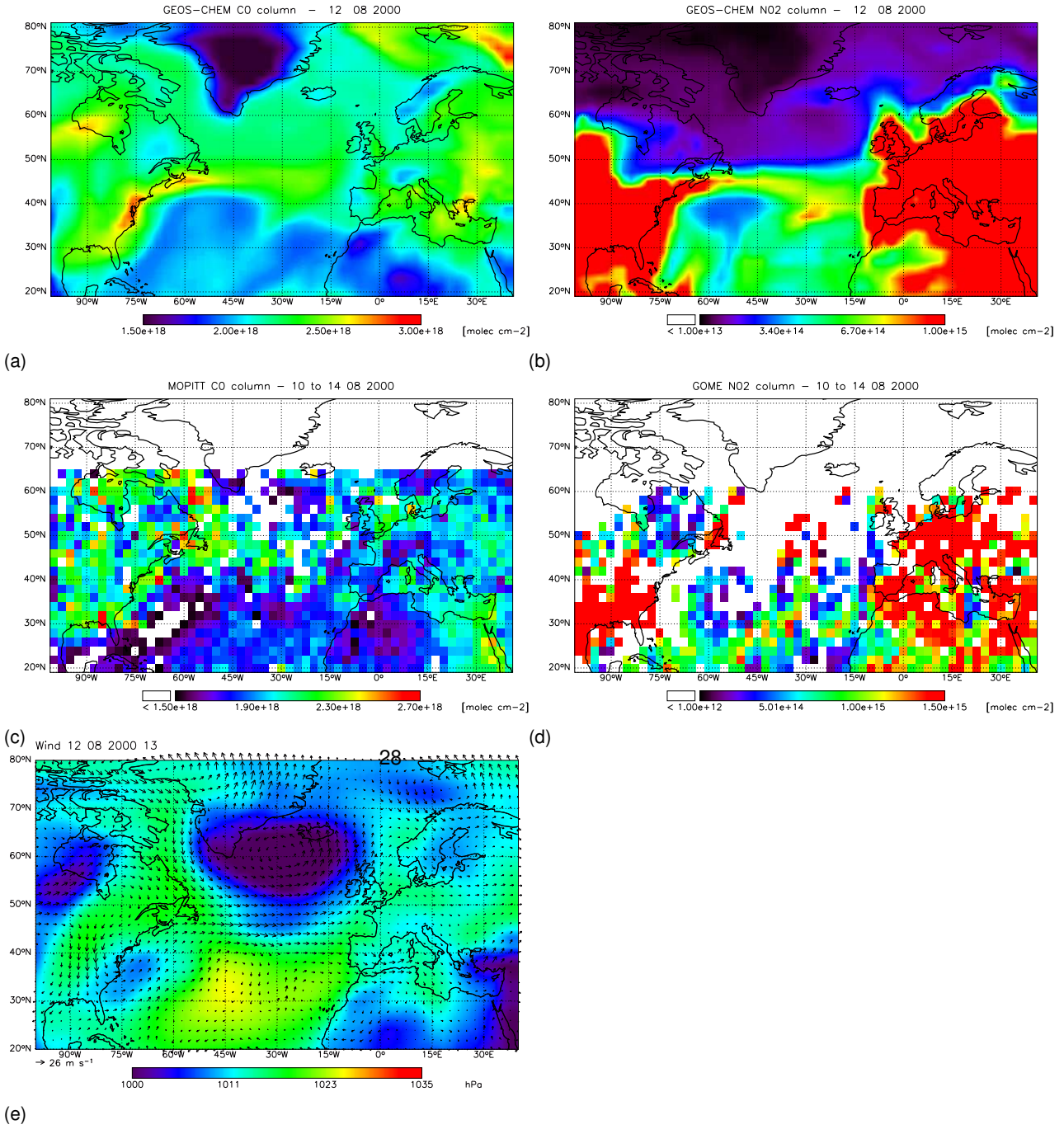

Fig. 4. Illustration of LRT9 event: **(a)** GEOS-CHEM CO column on 12 August, **(b)** GEOS-CHEM NO_2 column on 12 August, **(c)** MOPITT CO column on 10–14 August, **(d)** GOME NO_2 column on 10–14 August and **(e)** GEOS-CHEM mean sea level pressure and 500 hPa winds on 12 August.

associated with post-frontal outflow. The transport of that plume is taking place at the periphery of the Azores anticyclone and the plume is then turning anticyclonicaly, i.e. to the south, shortly before reaching Europe.

The fourth episode (LRT4) is observed in the period 5–9 July (Fig. 3). On 5 July a WCB reaches its highest altitude over the Gulf of Saint Lawrence (Li et al., 2005). A CO plume collocated with a NO_2 enhancement is apparent in GEOS-CHEM close to Greenland on 8 July (Figs. 3a, b).

Table 1. Summary of transatlantic episodes in summer 2000. Column 1: Episode number. Columns 2 and 3: Starting date and location, respectively, in North America. Column 4: Transport pathway over the North Atlantic; C stands for cyclone while Z stands for zonal transport. Columns 5 and 6: Date and place of arrival over Europe. Column 7: Plume classification based on emission types; A stands for anthropogenic while AB indicates a mix of anthropogenic and biomass burning emissions. Column 9: Availability of MOPITT and GOME data; "na" indicates that no observation is available for that episode. The episodes marked with a star are described in Sect. 4.

episode	North America		transatlantic	Europe		Plume	MOPITT
	Starting date	Location	transport	Arrival date	Location	origin	GOME
LRT1*	10–12 June	35–45° N	Z 40–50° N	13 June	UK Scandinavia	A	CO – yes NO_2 – yes
LRT2*	16–18 June	50–70° N accumulation	C 40–60° N mid Atlantic	21 June	Portugal/France Norway	A	CO – na NO_2 – na
LRT3	24–26 June	45–50° N accumulation	C 45° N mid Atlantic	29 June	Portugal France	A	CO – na NO_2 – na
LRT4*	5 July	45–50° N	C 60° N Greenland/Iceland	9 July	UK	A	CO – na NO_2 – na
LRT5	7–8 July	35–40° N	C 60° N Greenland/Iceland	13 July	UK Scandinavia	AB	CO – na NO_2 – na
LRT6	12–13 July	30–60° N	C 45° N mid Atlantic	21–22 July	Portugal Switzerland	A	CO – yes NO_2 – yes
LRT7	23–24 July	30–50° N	C 40° N mid Atlantic	30 July	Portugal Switzerland	A	CO – na NO_2 – na
LRT8	27–28 July	50–65° N accumulation	C 70° N Greenland/Iceland	5 August	Scandinavia	AB	CO – na NO_2 – na
LRT9*	9–12 August	45° N	Z 45° N	13–14 August	France	A	CO – yes NO_2 – partly

The cyclone and the plume travel toward Europe and reaches the UK on 9 July. The plume is subsequently transported by a new cyclone over the North Sea. As the Azores anticyclone occupies the entire North Atlantic (Fig. 3c), the transport can take place only north of 50° N. No MOPITT and GOME data are available to confirm that episode.

A zonal LRT event (LRT9) is observed in the period 9 to 14 August, which is similar to the LRT1 in terms of transport pathways. Both MOPITT CO and GOME NO_2 (Figs. 4c and d) show enhancements over the western Atlantic ocean between 40 and 50° N, even though only limited data are available for that period for GOME. The model also shows clear enhancements in the CO and NO_2 columns around 45° N (Figs. 4a and b), but a more detailed analysis shows that the outflow is restricted to the lower troposphere below 600 hPa.

5 Impact of LRT events on O_3 distributions over Europe

5.1 O_3 at Jungfraujoch

Figure 5 shows daily averaged O_3 concentrations observed at JFJ and simulated by GEOS-CHEM, along with the model North American, European and stratospheric O_3 contributions. The model underestimates the observed O_3 concentrations by 9 ppb (15%) on average over the summer and only captures some of the daily variability (r=0.37) (Table 2). The model predicts a stratospheric contribution of about 5 ppb (11%). Zanis et al. (2003) used observed $^7Be/^{10}Be$ ratio to derive an estimate of the contribution of stratospheric O_3 at JFJ, and found that on average over the summer 5 ppb (9%) originates from the stratosphere, with values ranging from almost 8 ppb in June to less than 3 ppb in August. Even though the stratospheric contribution remains more or less constant in our model over the course of the summer, we find similar values, indicating that the simulated stratospheric

Table 2. Monthly statistics for O_3 observed at Jungfraujoch (JFJ) and simulated by GEOS-CHEM. Monthly mean contributions are also shown, including North American (NA), European (EU) and stratospheric (ST) O_3. Numbers in parenthesis indicate the relative contribution to the simulated O_3. The remaining O_3 fraction (about 65%) includes contribution from anthropogenic emissions outside Europe and North America and contribution from natural sources. The last row indicates the stratospheric contribution as provided by Zanis et al. (2003).

	June 2000	July 2000	August 2000	Summer 2000
JFJ [ppb]	60.2	55.8	59.1	58.3
GEOS-CHEM [ppb]	50.3	48.8	47.5	48.9
Bias [ppb]	9.9 (16%)	7.0 (13%)	11.7 (20%)	8.7 (15%)
Corr. coeff. (r)	0.28	0.41	0.48	0.37
North American [ppb]	9.8 (20%)	9.7 (20%)	6.4 (14%)	8.7 (18%)
European [ppb]	3.3 (5%)	3.7 (7%)	3.0 (5%)	3.4 (6%)
Stratosphere [ppb]	5.9 (12%)	5.9 (12%)	4.0 (8%)	5.3 (11%)
Zanis ST mean [ppb]	7.7 (13%)	5.2 (9%)	2.7 (5%)	5.2 (9%)

contribution is likely to be reasonable for that season and period.

The model simulates an averaged European contribution of about 3 ppb (6%) at JFJ. As mentioned previously, the JFJ site is a mountain site which is frequently impacted by fronts and thermal lifting that bring planetary boundary layer (PBL) air with potentially high O_3 concentrations, especially in summer. We further analysed the air masses reaching JFJ using back trajectories calculated with the TRAJEC model of German Weather Service (Fay et al., 1995). This model uses a horizontal grid resolution of 15 km which should resolve local scale phenomena. We find that most of O_3 maxima observed at JFJ are associated with air masses originating from the European boundary layer below 800 hPa. Even though the GEOS-CHEM model shows a European contribution on those days, it is likely that the relatively coarse resolution used in our model prevents us from fully capturing the local phenomena and that the European contribution is underestimated at the JFJ site.

The average North American O_3 contribution is 9 ppb in summer 2000. Increases in the North American contribution above this mean value can be attributed to each of the previously discussed LRT events (see Fig. 5). During those events, we find that the North American O_3 at JFJ can reach up to 20 ppb (on an hourly base) and in some cases the enhancement is seen over several days. To confirm the origin of the long range transported air masses seen at JFJ, ten-day back trajectories were calculated with the LAGRANTO model (Wernli and Davies, 1997) which uses ECMWF (European Centre for Medium Range Weather Forcasts) meteorological fields with a spatial resolution of 1° and a temporal resolution of 6 h. Seven trajectories were calculated 4 times per day (i.e. arriving at 00:00, 06:00, 12:00, and 18:00 UTC), including one arriving at the JFJ site, 4 displaced by 0.5° in the horizontal and 2 displaced by 25 hPa in the vertical. We find that, for all the days for which GEOS-CHEM predicts a North American contribution greater than 10 ppb (i.e. 15–16 June, 23–24 June, 30 June–1 July, 8–9–10 July, 13–14–15 July, 26–27 July, 31 July, 6–7 August, 15 August), at least some of the air masses arriving at JFJ had a contact with the North American PBL i.e., they originated from or passed through the PBL of North America. In particular, the trajectory analysis indicates clearly a North American origin for the strongest episodes, e.g., 16 and 7 out off the daily 28 trajectories did originate from North America for LRT1 and LRT4, respectively, on average during the 2 or 3 days of the episodes. In contrast, for the days for which GEOS-CHEM predicts only a small North American contribution (less than 10 ppb), the back trajectory analysis indicates no contact with the North American boundary layer.

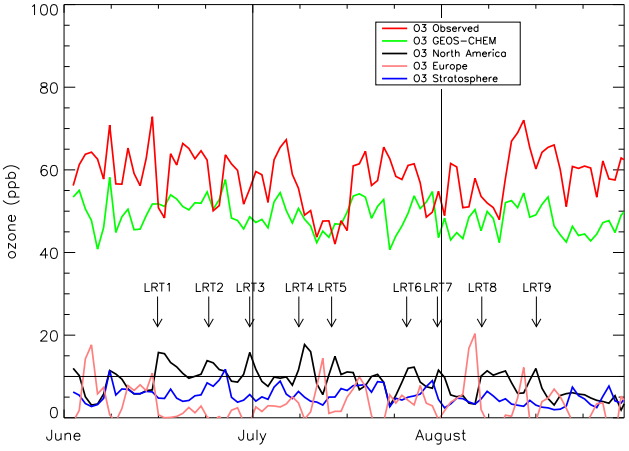

Fig. 5. Observed (red line) and simulated (green line) O_3 at JFJ in June–August 2000. Results from model sensitivity simulations are also shown, including the North American O_3 (black line), European O_3 (orange line) and stratospheric O_3 (blue line) contributions (see Sect. 3 for description of the sensitivity simulations).

During most of the events, a strong contribution from North American O_3 is associated with only little European O_3 and vice-versa. This reflects the successive occurrence of fair weather/stagnant conditions over Europe (leading to a stronger development of the boundary layer over Europe

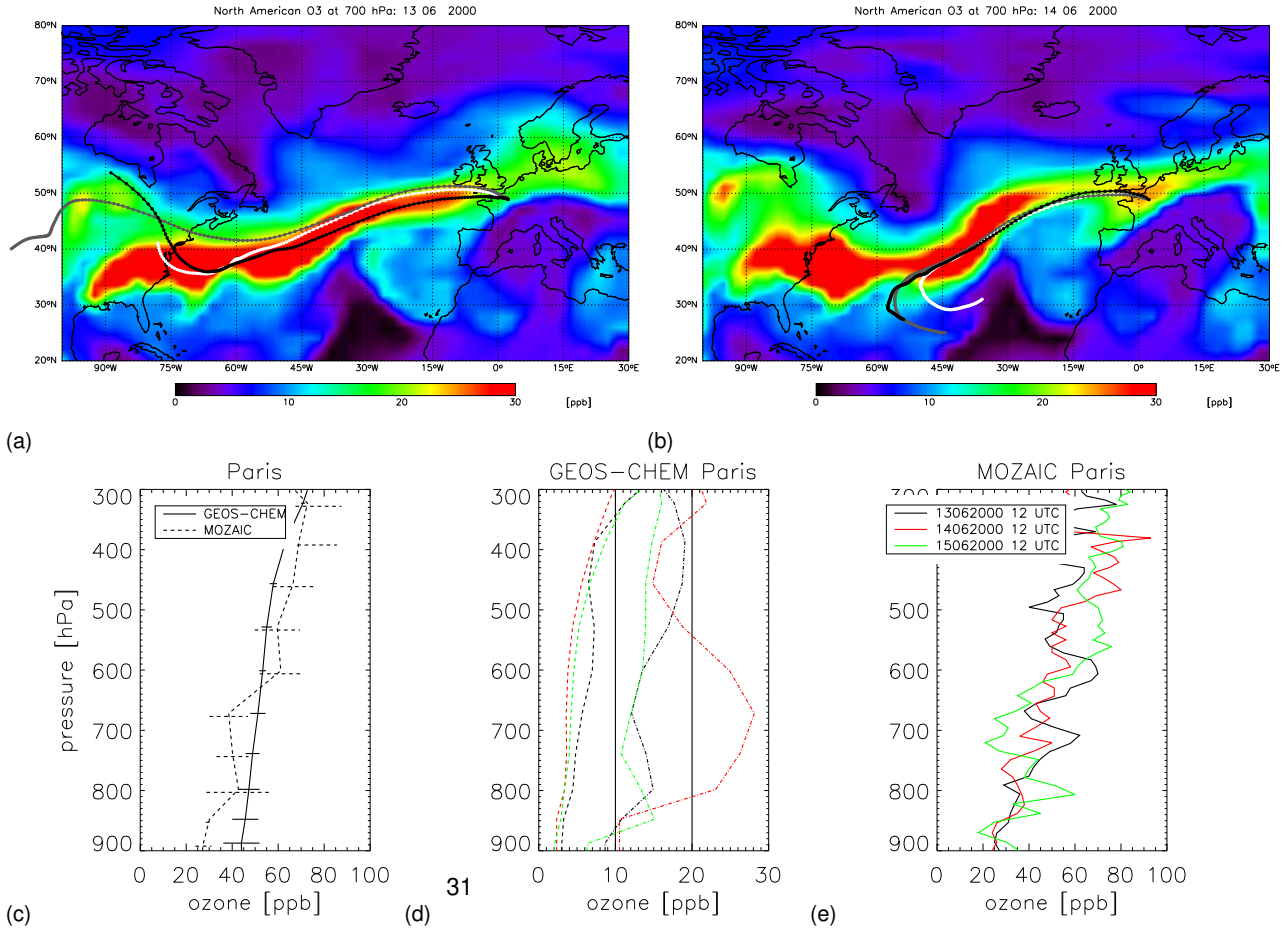

Fig. 6. LRT1: Daily mean North American O_3 at 700 hPa and MOZAIC back trajectories arriving in Paris at 12:00 UTC at 850 (white line), 700 (grey line) and 500 hPa (black line) on **(a)** 13 June and **(b)** 14 June. **(c)** O_3 averaged over the period 13–15 June (and standard deviation) at Paris from GEOS-CHEM (solid line) and MOZAIC (dashed line). MOZAIC data are interpolated on the GEOS-CHEM vertical levels. **(d)** North American O_3 (dash-dotted line) and stratospheric O_3 (dashed line) at Paris from GEOS-CHEM on 13–15 June. **(e)** MOZAIC profiles on 13–15 June.

that can even reach the JFJ site) and of perturbed weather conditions (associated with cyclone passages which bring North American pollution in some cases). The strongest LRT events are thus not associated with the highest levels in O_3 concentrations, which makes their identification in the O_3 observations difficult. This is different from what is reported by Huntrieser et al. (2005), who found that LRT events from North America can lead to enhanced daily O_3 concentrations at some Alpine sites. However, Huntrieser et al. (2005) investigated LRT events in the late fall season, when the European sources have a weaker influence on O_3 concentrations at mountain sites. Also, contrary to what is shown in Huntrieser et al. (2005), we do not find LRT events to be associated with enhanced CO concentrations at JFJ (not shown here), because of the much shorter lifetime of CO in the summertime than in the fall.

5.2 O_3 profiles over central Europe

In this section, we apply the GEOS-CHEM model to interpret O_3 MOZAIC profiles observed over central Europe during four representative LRT events. We show observed and simulated profiles over Paris even though the events are seen at Brussels and Frankfurt, at least in the model. We also use back trajectories at 850, 700 and 500 hPa available from the MOZAIC database (Simon et al., 2000) to trace back the origin of the air masses. The trajectory model uses the ECMWF meteorological data with a spatial resolution of 60 km and a temporal resolution of 6 h. The trajectories are 6.5-day backwards and are available each day at 12:00 UTC. High concentrations of NO_x close to airports and in general in the boundary layer close to large cities induce a strong titration of O_3, which may not be reflected in the model grid. Therefore vertical profiles are shown only above 900 hPa.

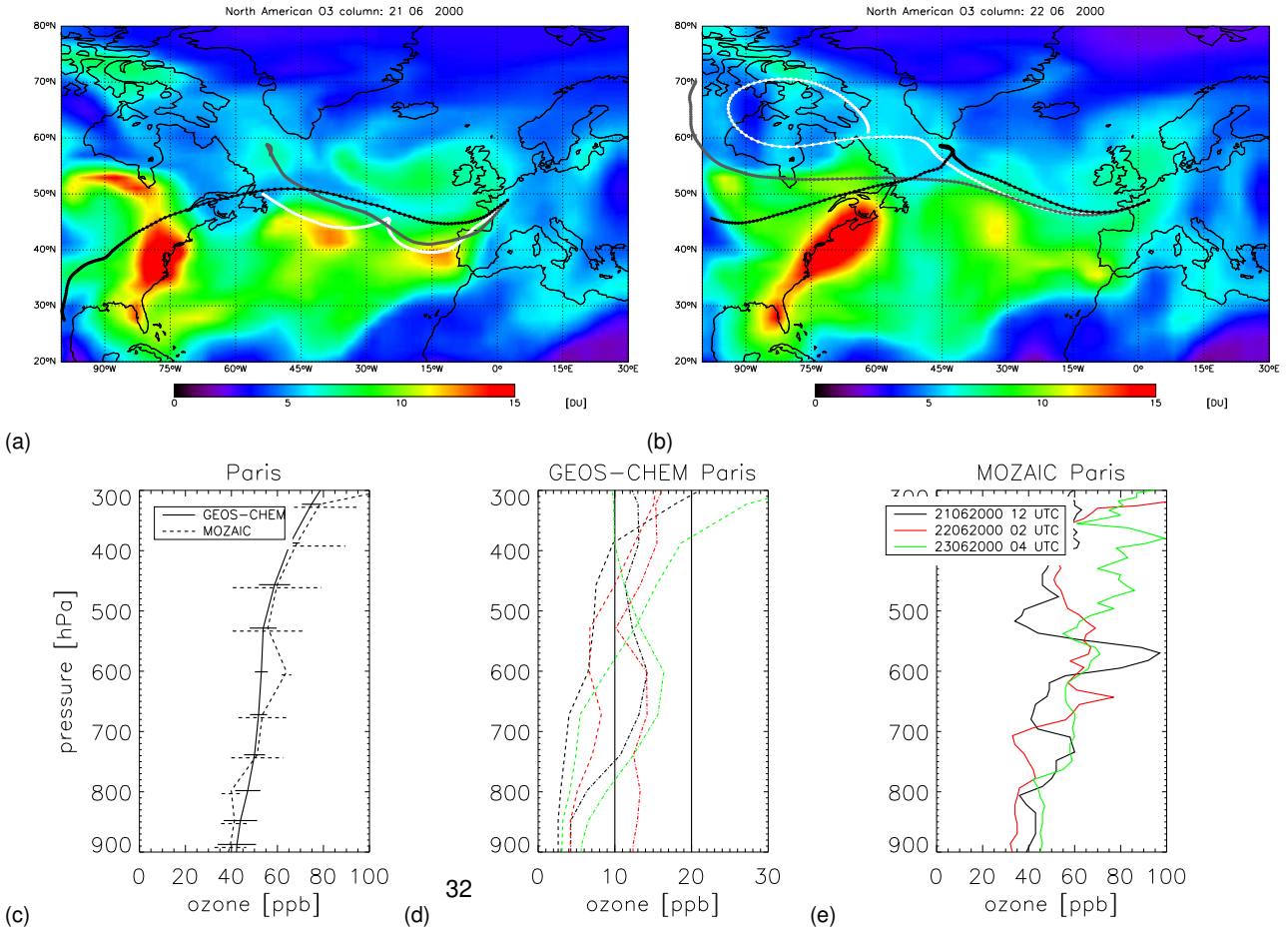

Fig. 7. LRT2: Daily mean North American O_3 tropospheric column and MOZAIC back trajectories arriving in Paris at 12:00 UTC at 850 (white line), 700 (grey line) and 500 hPa (black line) on **(a)** 21 June and **(b)** 22 June. **(c)** O_3 averaged over the period 21–23 June (and standard deviation) at Paris from GEOS-CHEM (solid line) and MOZAIC (dashed line). MOZAIC data are interpolated on the GEOS-CHEM vertical levels. **(d)** North American O_3 (dash-dotted line) and stratospheric O_3 (dashed line) at Paris from GEOS-CHEM on 21–23 June. **(e)** MOZAIC profiles on 21–23 June.

As expected, we find that the observed profiles result from a complex mixture of individual processes, including local photochemical activity, stratospheric intrusion and transport from North America. As further discussed in the following, the model cannot fully reproduce the observed daily variability, but nevertheless provides insight into the main processes which contribute to shape the observed profiles (e.g., stratospheric input versus LRT from North America).

The impact of the first event LRT1 is illustrated in Fig. 6 as the plume enters the European region on 13 June (Fig. 6a) and expands on 14 June (Fig. 6b). Figure 6c compares observed and simulated O_3 concentrations averaged over a 3-day period during the episode over Paris. The model reproduces the observations within 10–15 ppb above 900 hPa. The GEOS-CHEM sensitivity simulations indicate a strong enhancement in the North American contribution over a 3-day period (13–15 June) and especially on 14 June where it reaches up to 28 ppb at 700 hPa. The 6.5-day back trajectories arriving in Paris on 13 June (Fig. 6a) indicate that a fast transport took place across the Atlantic. This may explain the enhanced O_3 layers seen in the observed profiles at 700 hPa and 600 hPa on 13 June (Fig. 6b). On 14 June there is no clear enhancement in the observed O_3 profile (Fig. 6e) , and in fact the back trajectories arriving in Paris on 14 June seem to indicate that the air masses originate from the mid-Atlantic. This may suggest that the arrival of the North American plume into Europe is displaced in the GEOS-CHEM model by a few hours. During this episode, which is clearly the strongest one in summer 2000 according to the GEOS-CHEM model, the North American contribution reaches up to 28 ppb at around 700 hPa.

The LRT2 plume approaches Western Europe on 21 June as seen in Fig. 7a and further expands over central Europe on 22 June (Fig. 7b). The model compares relatively well to the

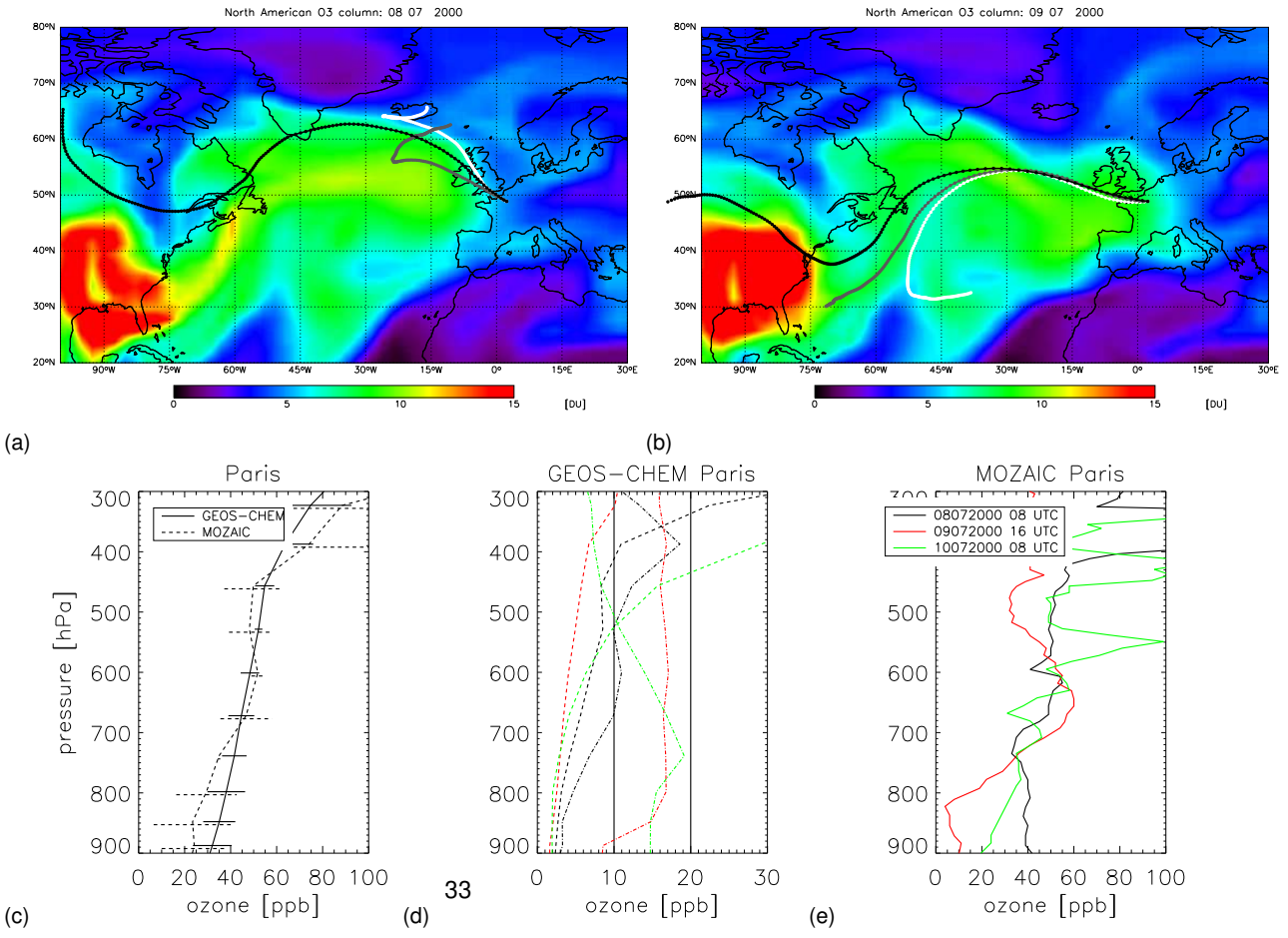

Fig. 8. LRT4: Daily mean North American O_3 tropospheric column and MOZAIC back trajectories arriving in Paris at 12:00 UTC at 850 (white line), 700 (grey line) and 500 hPa (black line) on **(a)** 8 July and **(b)** 9 July. **(c)** O_3 averaged over the period 8–10 July (and standard deviation) at Paris from GEOS-CHEM (solid line) and MOZAIC (dashed line). MOZAIC data are interpolated to the GEOS-CHEM vertical levels. **(d)** North American O_3 (dash-dotted line) and stratospheric O_3 (dashed line) at Paris from GEOS-CHEM on 8–10 July. **(e)** MOZAIC profiles on 8–10 July.

observations over the 3-day period (Fig. 7c). Sensitivity simulations by the GEOS-CHEM model indicate a broad North American contribution of about 15 ppb especially between 700 and 500 hPa over the 3-day period. The back trajectories confirm the North American origin of the air masses arriving at various altitudes (850, 700 and 500 hPa) over Paris, especially on 22 June (Fig. 7b). The observed profile on 21 June shows an enhanced O_3 layer of up to 95 ppb at 600 hPa which could be of North American origin (Fig. 7e). On 23 June an O_3 enhancement of about 25 ppb is also observed above 500 hPa, but this is likely of stratospheric origin according to the model sensitivity simulations (Fig. 7d).

The LTR4 plume reaches Europe at high altitude on 8 July (Fig. 8a) and spreads toward central Europe on 9 July (Fig. 8b). The averaged model profile is too high by about 10 ppb below 700 hPa and too low by about 10–20 ppb above 400 hPa (Fig. 8c). Examination of individual profiles and model sensitivity simulations show that the strong enhancements on 8 and 10 July above 400 hPa are likely associated with stratospheric input, which is represented qualitatively by the model. We find that the North American O_3 contributes substantially ($>$10 ppb) on 8 July at high altitude (above 500 hPa) and on 9 July over a broad altitude range in agreement with the back trajectories arriving in Paris. The observed profile on 9 July shows a broad layer centered around 750 hPa, which could be linked to the LRT event (Fig. 8e).

The LRT9 plume penetrates into continental Europe on 14 August (Fig. 9a and b). The model underestimates O_3 concentrations by about 10–20 ppb in the middle troposphere (Fig. 9c), as already mentioned in previous cases. In particular, the large enhancement observed at 500 hPa over the 3-day period is not well captured by the model. According to the model the large increase observed below 600 hPa at

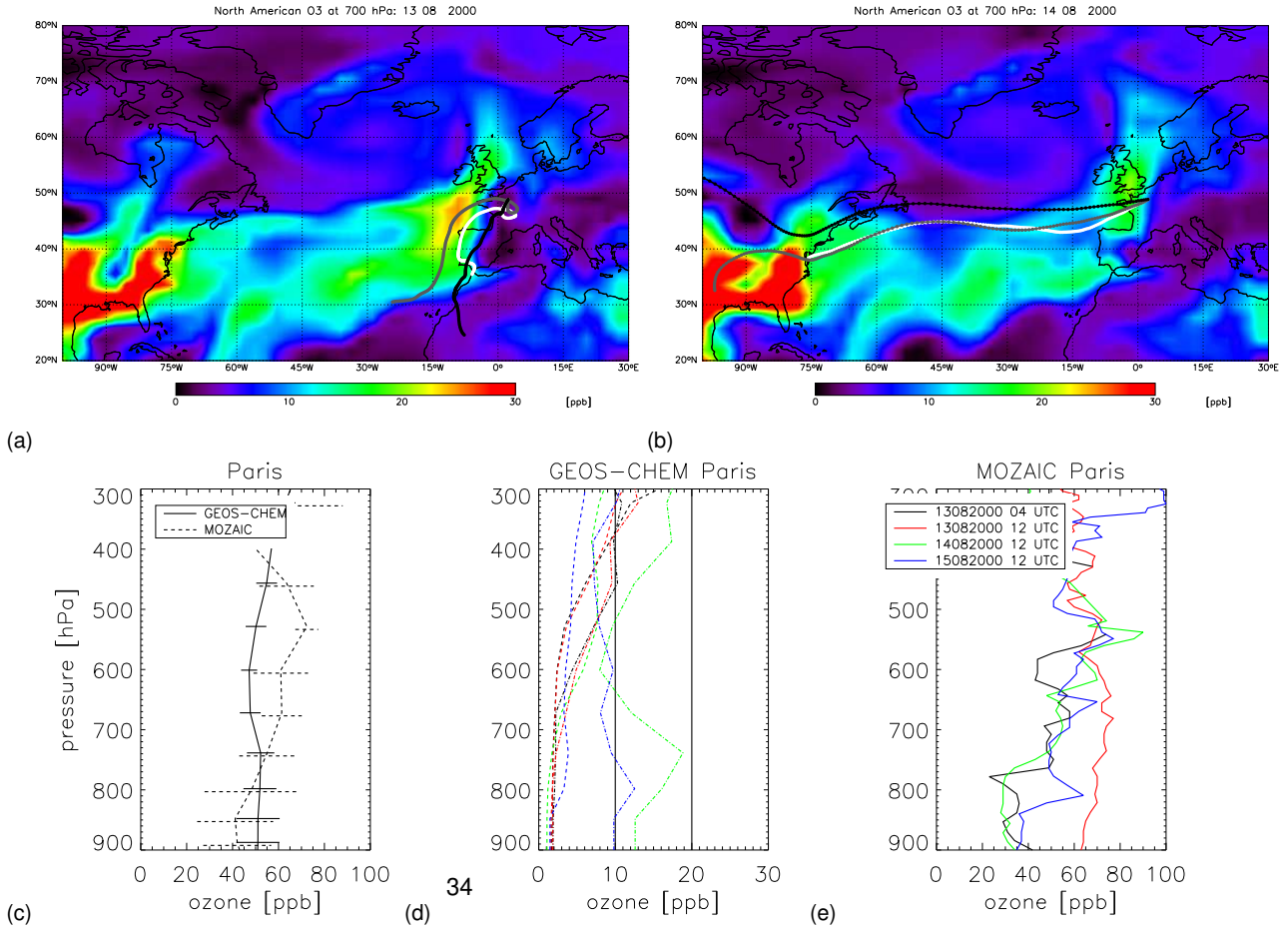

Fig. 9. LRT9: Daily mean North American O_3 at 700 hPa and MOZAIC back trajectories arriving at Paris at 12:00 UTC at 850 (white line), 700 (grey line) and 500 hPa (black line) on **(a)** 13 August and **(b)** 14 August. **(c)** O_3 averaged over the period 13–15 August (and standard deviation) at Paris from GEOS-CHEM (solid line) and MOZAIC (dashed line). MOZAIC data are interpolated on the GEOS-CHEM vertical levels. **(d)** North American O_3 (dash-dotted line) and stratospheric O_3 (dashed line) at Paris from GEOS-CHEM on 13–15 August. **(e)** MOZAIC profiles on 13–15 August.

12:00 UTC on 13 August is due to strong local O_3 production in an aging air mass, as also indicated by the back trajectories which recirculate around Paris. The model finds a large North American contribution (15 ppb) only on 14 August, in agreement with the back trajectories which clearly attribute a North American origin for the air masses detected over Paris on that day. The model predicts a substantial North American contribution (e.g. >10 ppb) only above 400 hPa and below 700 hPa and does not indicate any large contribution of stratospheric origin (Fig. 9d). Therefore the reasons for the large model underestimate seen between 600 and 500 hPa remains unclear.

A generalization of the approach used to quantify the impact of LRT events on the O_3 distribution over Europe is provided in Fig. 10, which shows the simulated North American O_3 contribution at 00:00 UTC and 12:00 UTC for each day of June, July and August 2000 over Paris, as an example (we found similar features over the other European cities Brussels and Frankfurt). The color dots correspond to the nine episodes and it is clear from Fig. 10 that the strongest episodes have been identified. The monthly mean profiles of North American O_3 present a broad maximum between 700 and 400 hPa and increase from about 5 ppb in the boundary layer to about 12 ppb in the middle and upper troposphere. The contribution in August is usually lower than in the two previous summer months. When examining individual events, we find that the North American contribution can reach up to 28 ppb (i.e. during LRT1). In the boundary layer (i.e. below 800 hPa), the North American contribution rarely exceeds 12 ppb. The impact of the zonal transport events (i.e. LRT1 and LRT9) which transport pollution at lower levels in general, is more clearly seen below 700 hPa while the events associated with cyclone are more likely to have an impact on the middle and upper-trospophere (i.e. above 600 hPa). Some

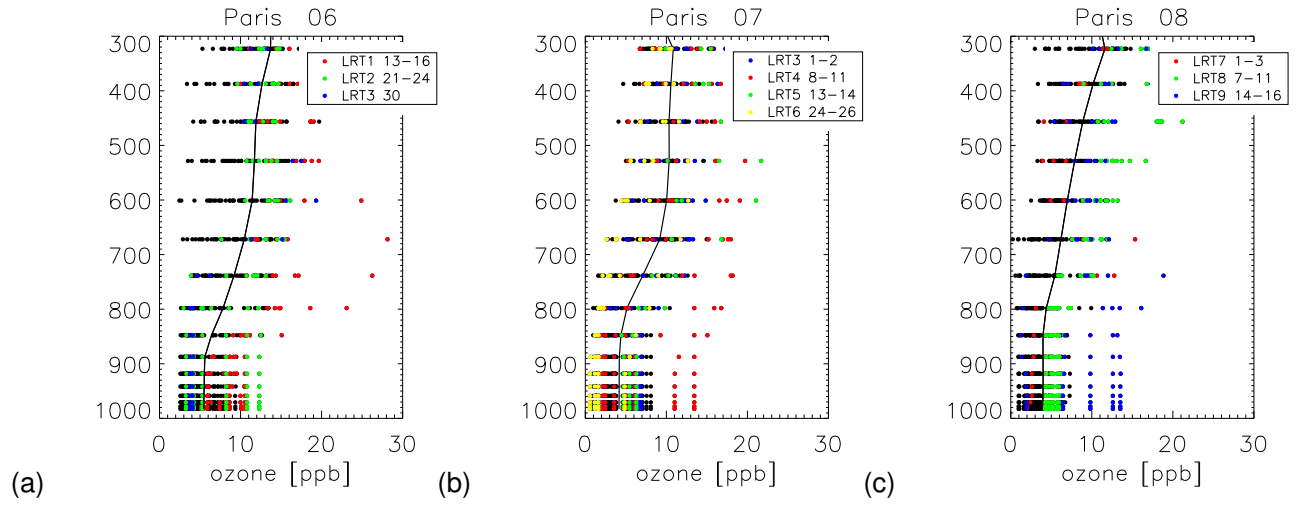

Fig. 10. North American O_3 contributions from GEOS-CHEM at Paris in **(a)** June 2000, **(b)** July 2000 and **(c)** August 2000. The solid lines correspond to the monthly averaged contributions and the dots correspond to instantaneous contributions at 00:00 and 12:00 UTC for each day of the month.

episodes, however, impact the O_3 distribution throughout the column (e.g., LRT4).

6 Summary and conclusions

In this paper, we examine LRT events from North America to Europe using the global chemical transport model GEOS-CHEM in conjunction with CO and NO_2 satellite observations by the MOPITT and GOME instruments, respectively. Then we use the model i) to analyse in-situ O_3 observations taken over central Europe, and ii) to quantify the impact of the LRT events on the O_3 distribution.

We find that the number and frequency of transatlantic LRT events depend strongly on the position and strength of the Azores anticyclone. After leaving North America, the plumes can either i) travel in the North American cyclones tracking northward and thus reach Europe at high latitudes; ii) be transported zonally between 40° and 55° N directly to Europe; iii) be incorporated into the Azores anticyclone and reach Europe at mid-latitudes; iv) never reach Europe (in the case of recirculation in the Azores anticyclone). Li et al. (2005a) reported 21 events of pollution export out of North America in summer 2000. We find that only 9 of them actually reach central Europe and have a substantial impact on the O_3 distributions. On average three episodes occur per month during the summer 2000 with the strongest ones being in June.

Comparison with observations at JFJ and from the MOZAIC program show that the model tends to underestimate O_3 concentrations in the middle troposphere. At the JFJ site, for example, simulated O_3 concentrations are too low by 9 ppb over the 3 months. As the stratospheric contribution is shown to be reasonable based on estimate of observed $^7Be/^{10}Be$ ratio, we suggest that this discrepancy results from an underestimate of the European contribution at JFJ. In addition, the model does not fully reproduce the daily variability observed at the JFJ and in the MOZAIC profiles. It is nevertheless possible to analyse the observed O_3 concentrations in light of model sensitivity simulations which provide the North American and stratospheric O_3 contributions.

During most of the 9 events, a strong contribution in North American O_3 at the JFJ is associated with only little European O_3 and vice-versa. We find that, in summer, a substantial North American contribution over Europe does not always result in pronounced O_3 enhancements in the observations. The air mass origin found in the GEOS-CHEM model is clearly confirmed by back trajectory analysis. Analysis of MOZAIC profiles reveals that the impact of LRT events is seen at a very large altitude range and with varying strength over the course of the summer. The summer mean North American O_3 contributions range from 3–5 ppb (7–11%) in the PBL and 10–13 ppb (18–23%) in the middle and upper troposphere based on GEOS-CHEM sensitivity simulations. During particular episodes North American sources can result in O_3 enhancements up to 25–28 ppb in the layer between 800–600 hPa and 10–12 ppb in the boundary layer. LRT events associated with transient cyclones tend to impact Europe in the middle and upper troposphere (i.e., above 600 hPa) while those associated with zonal transport impact more strongly the lower troposphere (i.e., below 600 hPa).

Our analysis reveals the difficulty of assessing the impact of LRT events on European O_3 distributions based on the sole use of O_3 observations. This study also indicates that a clear assessment of the impact of LRT on a given region can only be achieved through a dense network of measurement sites that provide vertical profiles with a high temporal resolution.

In addition, further work will be needed to investigate to what extent our results are specific to the year 2000.

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