

## Transatlantic transport of pollution and its effects on surface ozone in Europe and North America

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Received 24 October 2001; revised 2 December 2001; accepted 16 December 2001; published 3 July 2002.

[1] We examine the transatlantic transport of anthropogenic ozone and its impact on surface ozone in Europe and North America by using a 5-year (1993–1997) simulation with the GEOS-CHEM global three-dimensional model of tropospheric chemistry. Long-term time series of ozone and CO at Mace Head (Ireland) and Sable Island (Canada) are used to evaluate transatlantic transport in the model. North American anthropogenic emissions contribute on average 5 ppbv to surface ozone at Mace Head, and up to 10–20 ppbv during transatlantic transport events, which are forerunners of broader events in Europe. These events are associated with low-level westerly flow driven by an intense Icelandic low between Iceland and the British Isles. North American influence on ozone at Mace Head is strongly correlated with the North Atlantic Oscillation (NAO), implying that the NAO index can be used to forecast transatlantic transport of North American pollution to Europe. European anthropogenic emissions contribute on average less than 2 ppbv to surface ozone at Sable Island but up to 5–10 ppbv during transatlantic transport events. These events are associated with low-level easterly flow established by anomalous low pressure at 45°N over the North Atlantic. North American anthropogenic emissions enhance surface ozone in continental Europe by 2–4 ppbv on average in summer and by 5–10 ppbv during transatlantic transport events; transport in the boundary layer and subsidence from the free troposphere are both important mechanisms. We find in the model that 20% of the violations of the European Council ozone standard (55 ppbv, 8-hour average) in the summer of 1997 over Europe would not have occurred in the absence of anthropogenic emissions from North America. North American influence on surface ozone in Europe is particularly strong at the thresholds used for the European standards (55–65 ppbv). *INDEX TERMS*: 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS*: transatlantic transport, pollution, ozone, NAO index

### 1. Introduction

[2] There is presently much interest in intercontinental transport of ozone and aerosol pollution as a factor to be considered in the design of regional air quality control strategies [National Academy of Sciences, 2001; Committee

on Environment and Natural Resources, 2001]. Recent studies have indicated that transpacific transport of Asian pollution affects North America [Jacob *et al.*, 1999; Jaffe *et al.*, 1999; Wilkening *et al.*, 2000; Yienger *et al.*, 2000], transatlantic transport of North American pollution affects Europe [Derwent *et al.*, 1998; Ryall *et al.*, 1998; Stohl and Trickl, 1999], and transeurasian transport of European pollution affects Asia (H. Liu *et al.*, Sources of tropospheric ozone along the Asian Pacific Rim: An analysis of ozonesonde observations, submitted to *Journal of Geophysical Research*, 2001) (hereinafter referred to as Liu *et al.*, submitted manuscript, 2001). The magnitudes of these effects and the need to include them in air quality control strategies are still highly uncertain. In a global three-dimensional (3-D) model study, Fiore *et al.* [2002] found that anthropogenic sources in Asia and Europe enhance surface ozone in the United States by 3–7 ppbv in summer, with maximum effect under moderately polluted conditions (50–70 ppbv).

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[3] In this work, we quantify the transatlantic transport of ozone pollution and its effect on surface ozone in Europe and North America by using a 5-year (1993–1997) global 3-D model simulation evaluated with coastal observations on both sides of the North Atlantic. The Mace Head site (53°N, 10°W) on the west coast of Ireland is of particular interest. It receives westerly flow from the North Atlantic and is not impacted by local pollution sources [Jennings *et al.*, 1991]. Long-term measurements of ozone, carbon monoxide (CO), black carbon, and chlorofluorocarbons (CFCs) at Mace Head have been used to estimate transatlantic North American influence [Jennings *et al.*, 1996; Derwent *et al.*, 1998; Ryall *et al.*, 1998; Forster *et al.*, 2001]. Carbon monoxide is directly emitted by combustion sources and has a lifetime of 1–3 months in the atmosphere; therefore it is a sensitive tracer for long-range transport of pollution. Jennings *et al.* [1996] used air back-trajectories to indicate a North American origin for an event of weakly enhanced CO (30 ppbv above background) observed in the spring of 1992 at Mace Head.

[4] In the case of ozone, the complexity of sources and the short lifetime (days to weeks) make it difficult to quantify, or even identify, a transatlantic transport influence at the surface from simple analysis of observations [Fehsenfeld *et al.*, 1996a; Derwent *et al.*, 1998]. Stohl and Trickl [1999] identified by trajectory analysis an episode of long-range transport of North American ozone pollution in the upper over Europe following uplift by a warm conveyor belt (WCB) [Browning, 1999; Cooper *et al.*, 2001] over the western North Atlantic. As pointed out by Stohl [2001] and Stohl *et al.* [2002], such long-range transport of ozone pollution is far more efficient in the free troposphere than near the surface because of stronger winds and a longer ozone lifetime.

[5] Even though the influence of transatlantic pollution transport on surface ozone in Europe or North America may be too weak to be easily detected from observations, it is still highly relevant for regional air pollution. A transatlantic pollution enhancement of only a few ppbv would make the achievement of ozone air quality standards in Europe significantly more difficult [Jonson *et al.*, 2001]. Global 3-D models of tropospheric ozone-NO<sub>x</sub>-hydrocarbon chemistry offer a tool for quantifying such enhancements, provided that they are carefully evaluated for the purpose.

[6] We use the GEOS-CHEM global model [Bey *et al.*, 2001a] for our 5-year simulation. The model is driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Data Assimilation Office (DAO) [Schubert *et al.*, 1993]. Evaluations of the GEOS-CHEM simulation of tropospheric O<sub>3</sub>-NO<sub>x</sub>-hydrocarbon chemistry for different regions of the world have been presented in a number of papers [Bey *et al.*, 2001a, 2001b; Fiore *et al.*, 2002; Li *et al.*, 2001a, 2001b; Palmer *et al.*, 2001; Chandra *et al.*, 2002; Martin *et al.*, 2002a, 2002b; Liu *et al.*, submitted manuscript, 2001]. Fiore *et al.* [2002] reported detailed comparisons with observations for ozone and its precursors in the United States. Here we focus our model evaluation on observed time series of ozone and CO in 1993–1997 at Mace Head and Sable Island (44°N, 60°W), off the east coast of Canada. Measurements of ozone and CO at Mace Head are part of the Global Atmospheric Gases Experiment (GAGE) and later the Advanced GAGE (AGAGE) program [Simmonds *et al.*, 1997; Prinn *et al.*,

2000]. Measurements of ozone and CO at Sable Island are part of the North Atlantic Regional Experiment (NARE) and have been used to monitor North American continental outflow to the North Atlantic [Fehsenfeld *et al.*, 1996a, 1996b; Parrish *et al.*, 1993, 1998].

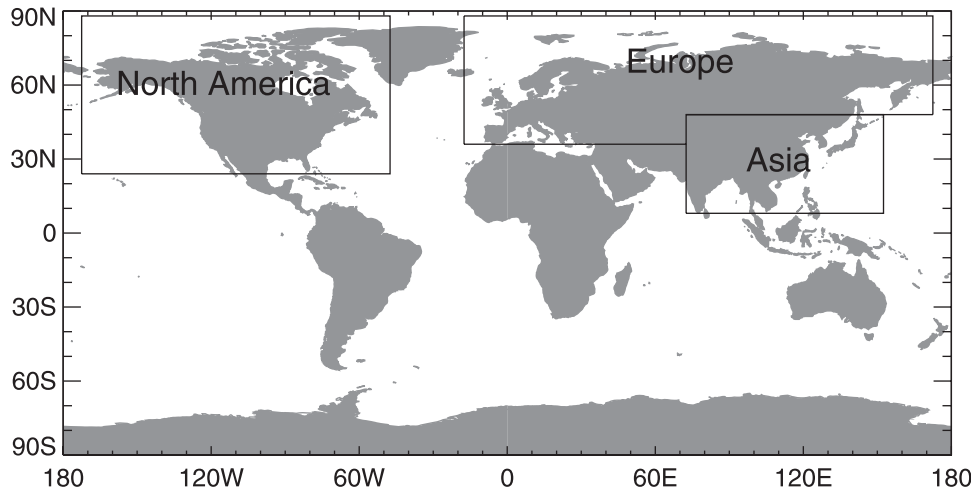
[7] We briefly describe the model simulation in section 2, including the tagged tracer and sensitivity analyses used to diagnose transatlantic transport of pollution. Transatlantic transport pathways are examined in section 3. In section 4 we analyze and interpret the model simulation of the observed time series of ozone and CO at the three North Atlantic sites. We quantify the impact of transatlantic pollution transport on surface ozone in Europe and North America in section 5. Summary and conclusions are given in section 6.

## 2. Model Simulation

[8] A detailed description of the GEOS-CHEM model for tropospheric O<sub>3</sub>-NO<sub>x</sub>-hydrocarbon chemistry is given by Bey *et al.* [2001a]. We use here model version 4.16 (available from Harvard University at <http://www-as.harvard.edu/chemistry/trop/geos>) applied to a continuous 5-year record (1993–1997) of GEOS assimilated meteorological observations with 3- or 6-hour temporal resolution depending on the variable and 2° latitude by 2.5° longitude horizontal resolution. The GEOS assimilation system is GEOS-1 for 1993–1995 and GEOS-STRAT for 1996–1997. Meteorological fields are provided on a sigma coordinate with 20 vertical levels up to 10 hPa in GEOS-1 and 46 up to 0.1 hPa in GEOS-STRAT. For computational expediency we degrade the horizontal resolution to 4° × 5° and merge the vertical layers above the lower stratosphere in the 1996–1997 GEOS-STRAT fields. There are typically 14–17 tropospheric vertical layers in the model. The lowest 3 layers extend to 100-m, 600-m, and 1-km altitude for a column based at sea level. The 5-year simulation is initialized on 1 January 1993 after a 1-year spin-up using meteorological fields from 1993.

[9] Anthropogenic emissions are specified using a base emission inventory for 1985 described by Wang *et al.* [1998a]. The base emission inventory is then scaled to specific years using national emission inventories and economic data [Bey *et al.*, 2001a]. The current model version includes updated CO emissions from Duncan and Logan [2001], and an improved emission inventory for biomass burning that includes interannual variability determined from satellite observations (B. N. Duncan *et al.*, Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, submitted to *Journal of Geophysical Research*, 2002). Ship emissions of NO<sub>x</sub> are from Benkovitz *et al.* [1996]. A sensitivity simulation using the much higher ship emission estimate of Corbett *et al.* [1999] yields NO<sub>x</sub> concentrations over the North Atlantic that are too high compared to aircraft observations, as previously reported in the global 3-D model study of Kasibhatla *et al.* [2000].

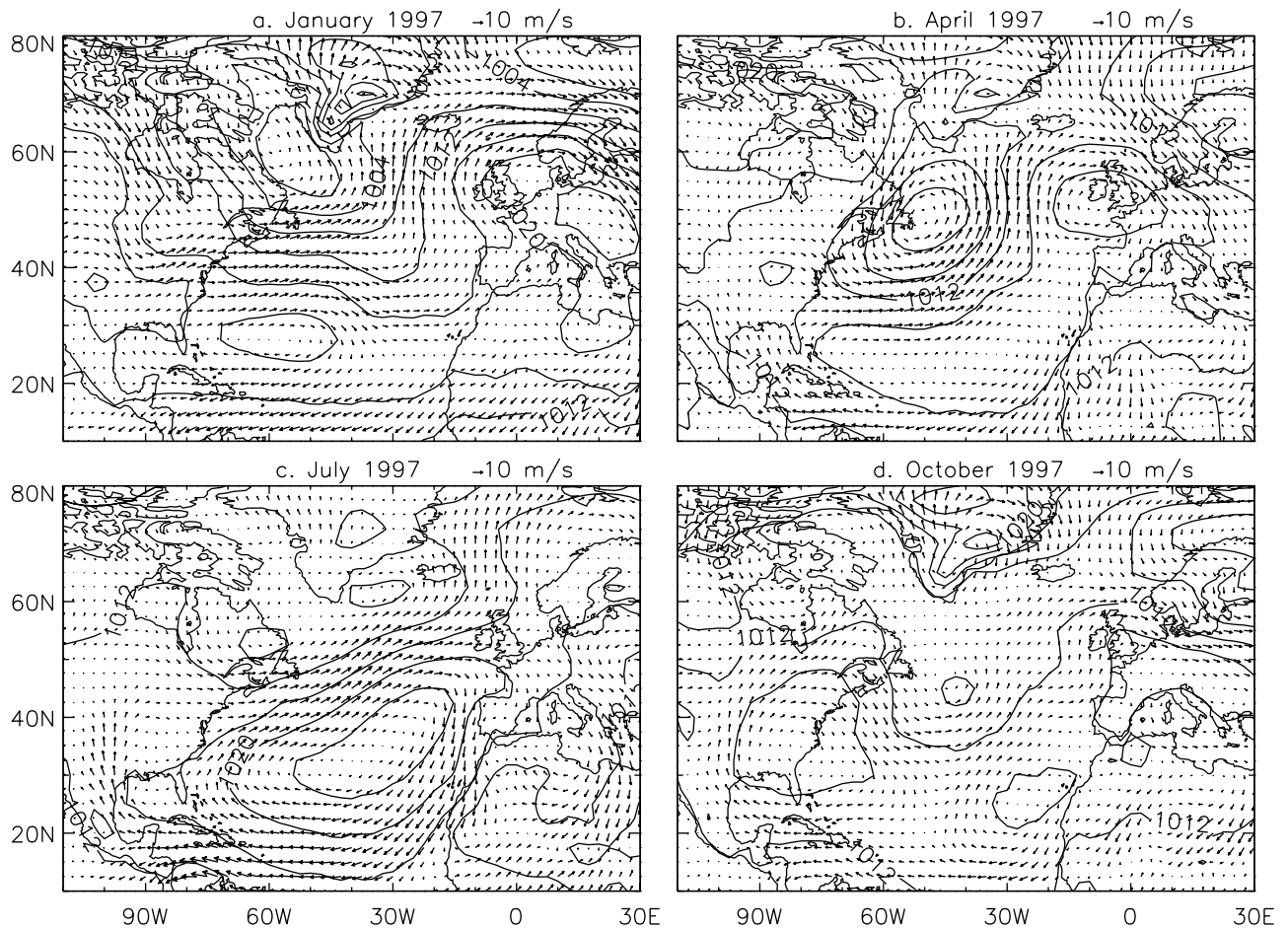
[10] We will present sensitivity simulations with anthropogenic emissions in North America, Europe, or Asia shut off to quantify their contributions to intercontinental transport. The sensitivity simulations are conducted from January to August 1997; the first 5 months are used for initialization and we focus our analysis on the summer of 1997. Summer is when ozone pollution is usually of greatest concern.



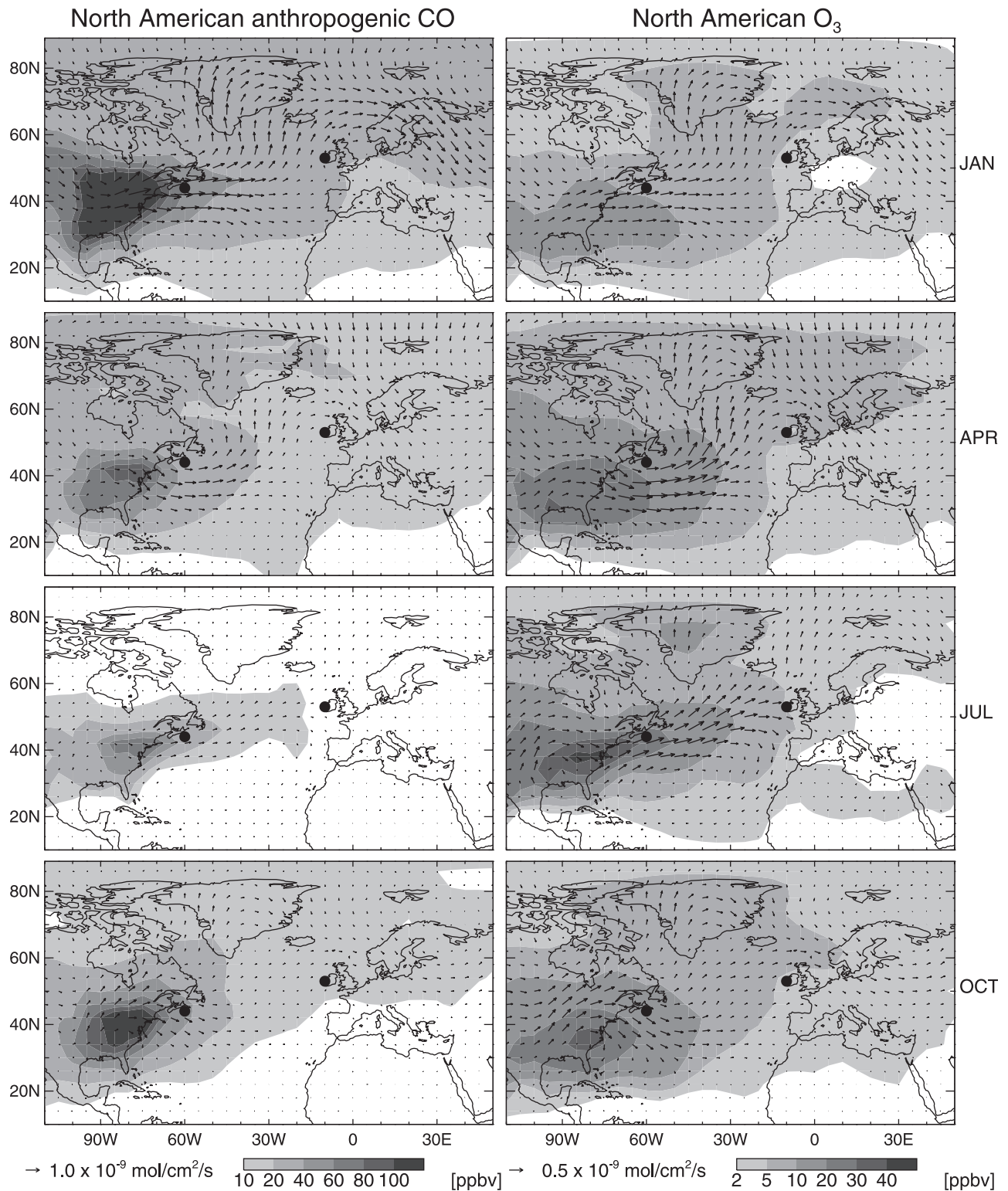
**Figure 1.** Source regions used for the tagged ozone and CO simulations.

[11] To gain a longer perspective on intercontinental transport, we also repeat the 5-year standard simulation using tagged CO and ozone tracers as a computationally expedient method for source attribution [Wang *et al.*, 1998b; Bey *et al.*, 2001b; Staudt *et al.*, 2001]. The tagged CO simulation separates contributions by source types [Bey *et al.*, 2001b; Staudt *et al.*, 2001] including anthro-

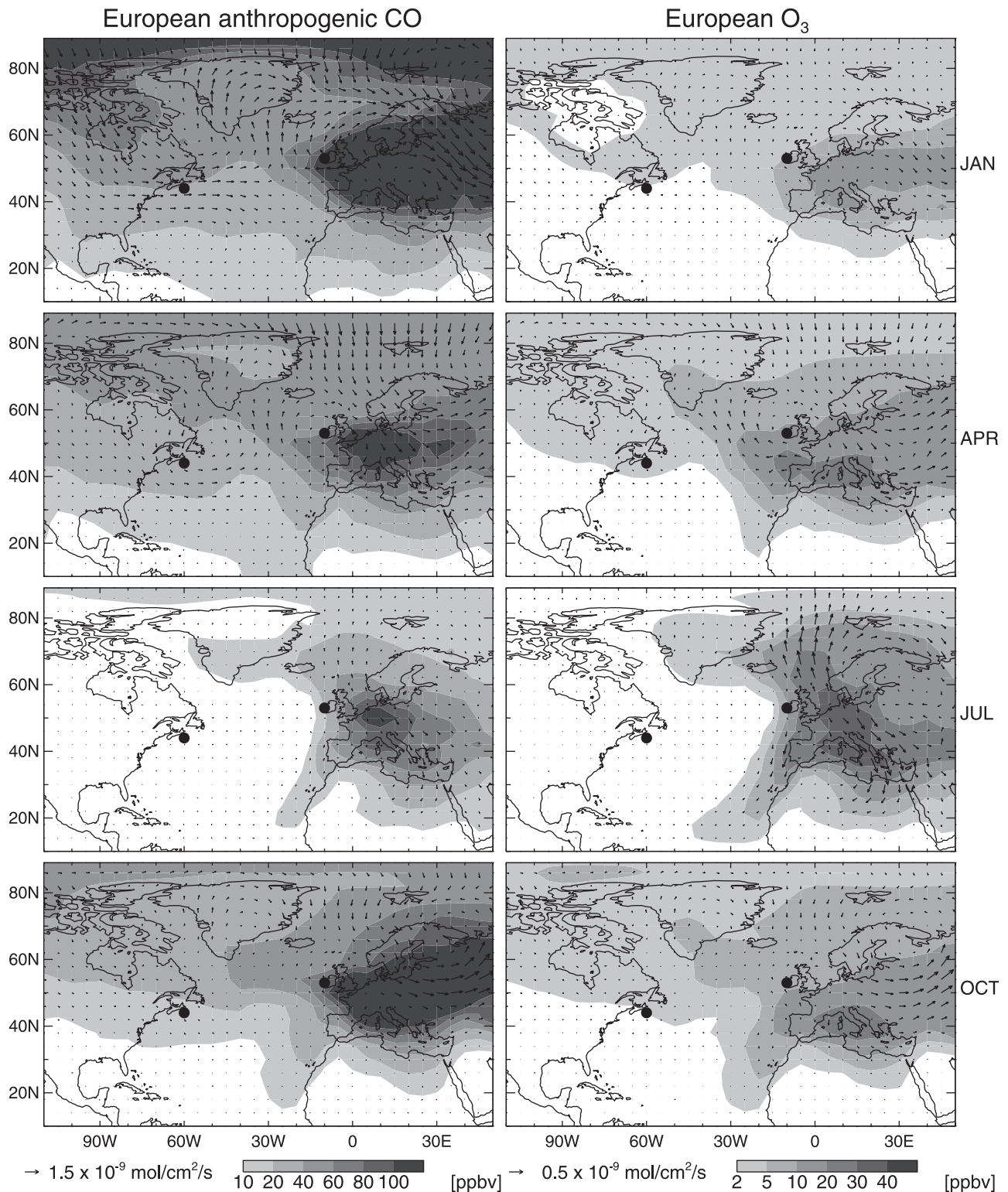
pogenic emissions in North America, Europe, and Asia (Figure 1). Here we define Europe to include eastern Russia, but emissions in eastern Russia are very small and inconsequential for the analysis that follows. Loss of the tagged CO tracers is computed using archived daily 3-D fields of OH from the standard full-chemistry simulation.



**Figure 2.** Monthly average GEOS sea level pressure and 925-hPa winds for (a) January, (b) April, (c) July, and (d) October 1997.



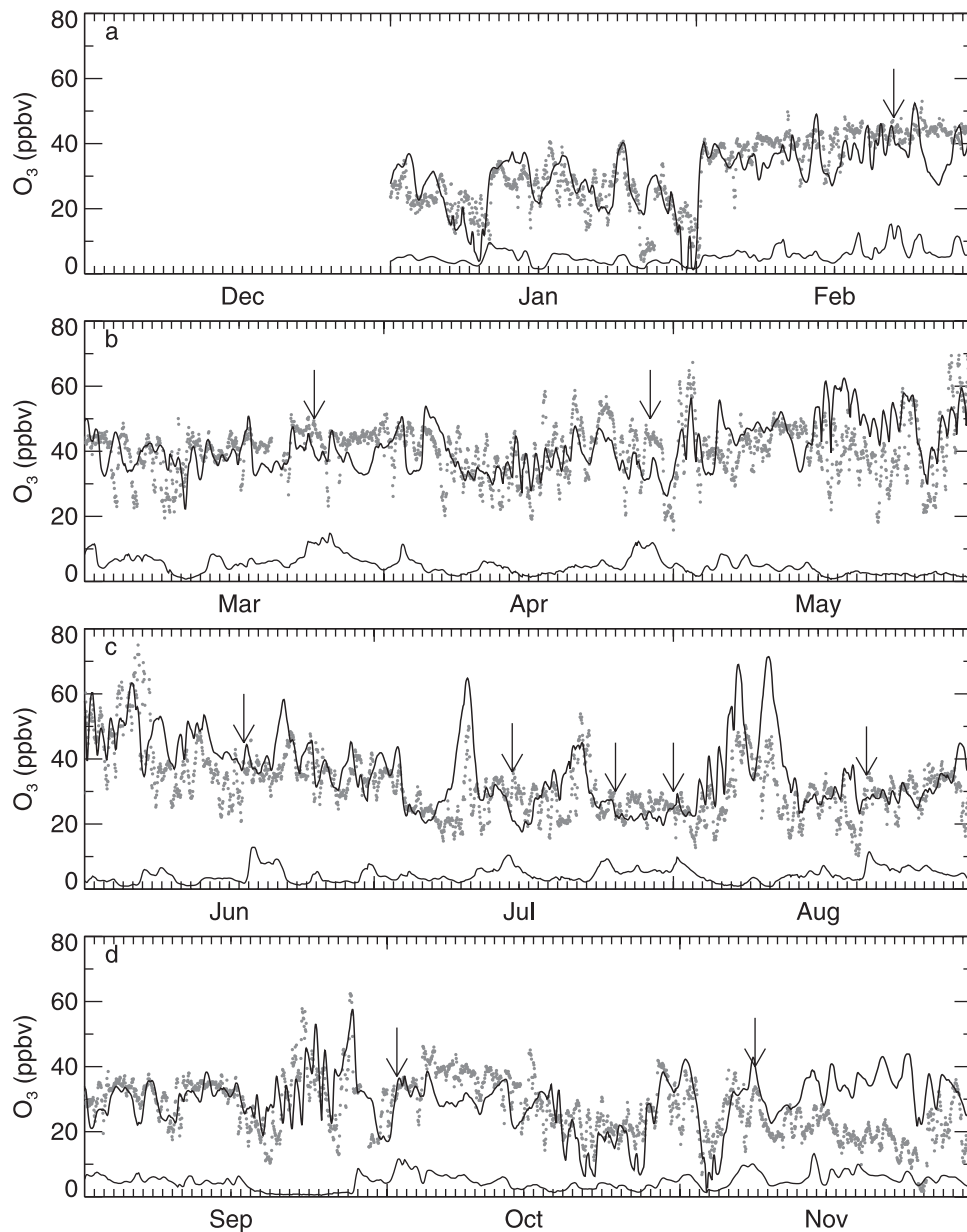
**Figure 3.** Simulated monthly average fluxes at 0–3-km altitude of (left) North American anthropogenic CO and (right) North American ozone for January, April, July, and October 1997. Contours are concentrations in the lowest model layer (0–100 m above the surface). Solid circles indicate the locations of Mace Head and Sable Island. See color version of this figure at back of this issue.



**Figure 4.** Simulated monthly average fluxes at 0–3-km altitude of (left) European anthropogenic CO and (right) European ozone for January, April, July, and October 1997. Contours are concentrations in the lowest model layer (0–100 m above the surface). Solid circles indicate the locations of Mace Head and Sable Island. See color version of this figure at back of this issue.

[12] In the case of ozone, we use archived daily 3-D fields of odd oxygen ( $\text{O}_x = \text{O}_3 + \text{NO}_2 + \text{NO}_3 + \text{HNO}_3 + \text{PAN} + \text{H}_2\text{O}_4 + \text{N}_2\text{O}_5$ ) production rates and loss frequencies from the standard full-chemistry simulation to trans-

port separate  $\text{O}_x$  tracers originating from the lower troposphere (below 700 hPa) over different continents (Figure 1), the lower troposphere over the oceans, the middle troposphere (400–700 hPa), the upper troposphere

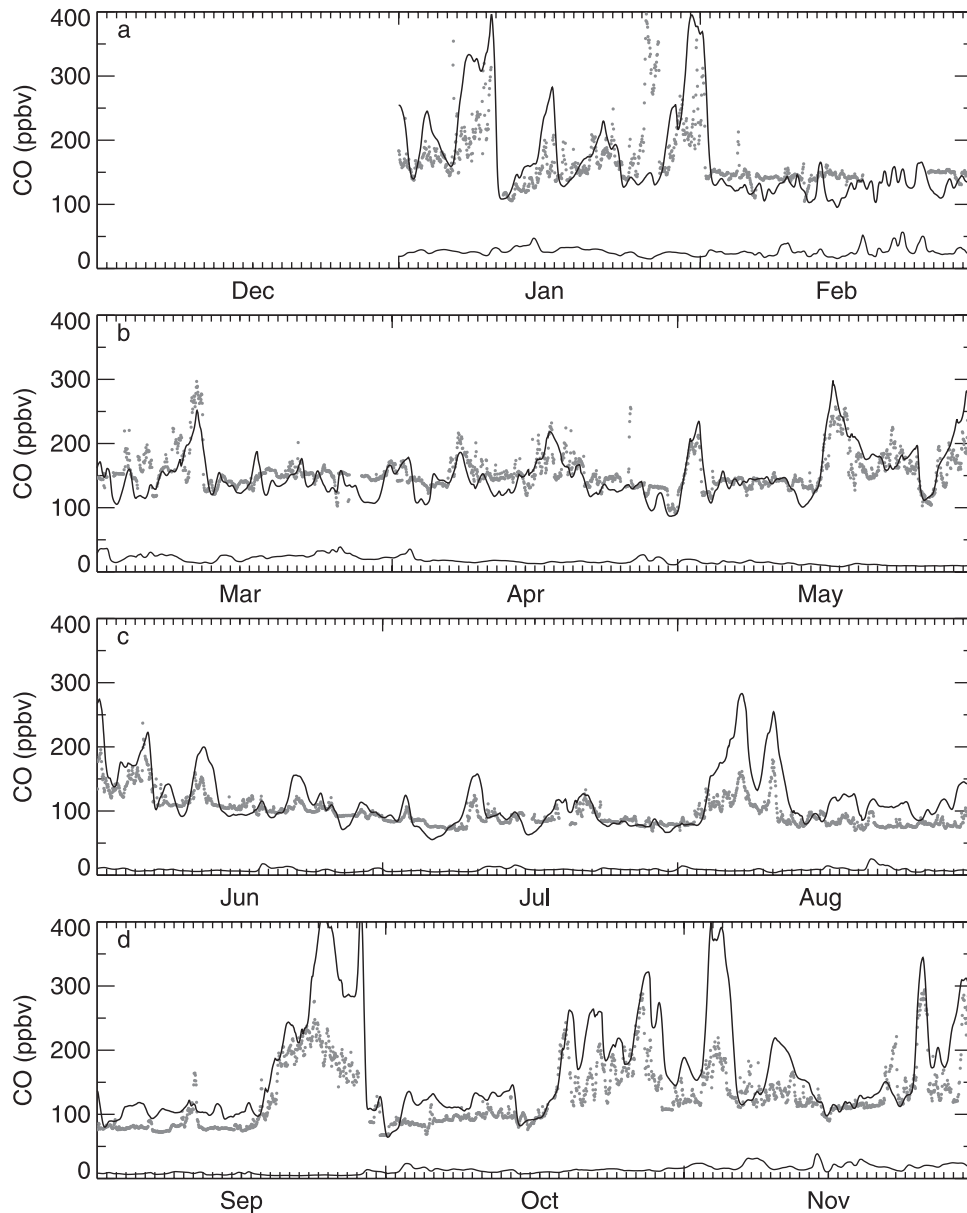


**Figure 5.** (a–d) Hourly average surface concentrations of ozone and for January–November 1997 at Mace Head ( $53^{\circ}\text{N}$ ,  $10^{\circ}\text{W}$ ) (gray dots, observations; thick lines, model). Contributions from North American ozone in the model are shown as thin lines. The arrows indicate major transatlantic transport events for ozone.

(400 hPa-tropopause), and the stratosphere. The tracers are removed by chemical loss and dry deposition at the same frequencies as those for total  $O_x$  in the standard simulation. By summing the concentrations of all the tracers, we reproduce closely the total  $O_x$  concentrations in the standard full-chemistry simulation. Since ozone typically accounts for over 95% of  $O_x$ , we will refer to the tagged  $O_x$  as tagged ozone in what follows for the sake of clarity. We will further refer to the tagged ozone tracer produced in the lower troposphere over North America as “North American ozone,” and similarly for the other source regions of Figure 1.

[13] Interpretation of results for tagged ozone tracers such as “North American ozone” requires caution. First,

“North American ozone” includes contributions from both anthropogenic and natural production within the continental lower troposphere, but does not include the contribution from production outside the continental lower troposphere driven by precursors emitted from North America. Second, because of the nonlinear chemistry involved in ozone production, the decrease of ozone in a simulation with North American sources shut off is less than the North American contribution to total ozone in the standard simulation. We find in practice that the contributions to surface ozone at Mace Head and Sable Island from the tagged North American and European ozone tracers replicate closely the results from the sensi-



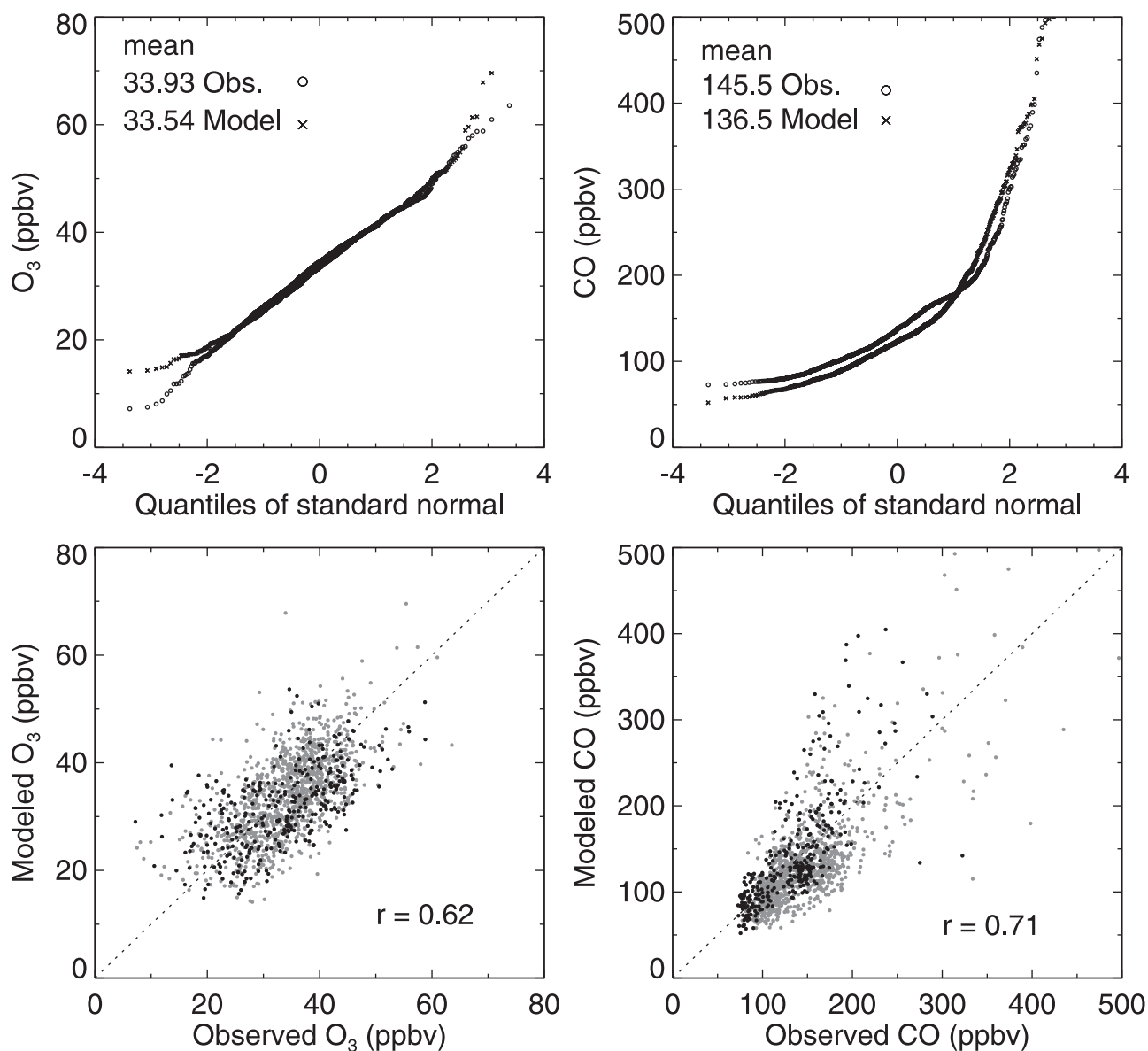
**Figure 6.** (a–d) Hourly average surface concentrations of CO for January–November 1997 at Mace Head ( $53^{\circ}\text{N}$ ,  $10^{\circ}\text{W}$ ) (gray dots, observations; thick lines, model). Contributions from North American CO in the model are shown as thin lines.

tivity simulations with anthropogenic sources in these regions shut off (see section 5). This result must reflect some compensation between nonlinear response and contributions from biogenic sources to ozone production in North America and Europe. It does imply that the tagged ozone tracer simulation offers a good measure of anthropogenic influence from different continents. Using the tagged ozone and CO tracers, we can conveniently probe the seasonal and interannual variability of transatlantic transport of pollution in the 5-year simulation.

### 3. Pathways for Transatlantic Transport

[14] The circulation over the North Atlantic is characterized by three main features: (1) the Icelandic low, (2) the

semipermanent Bermuda-Azores high, and (3) the trade wind region south of the Bermuda-Azores high [Tucker and Barry, 1984]. Figure 2 shows monthly average GEOS sea level pressures and 925-hPa winds for January, April, July, and October 1997. The winds are strongest along the storm track off the east coast of North America over the western North Atlantic. Over eastern North America between  $30^{\circ}$ – $60^{\circ}\text{N}$ , the westerly winds extend to the surface essentially year-round. This allows transport in the lower troposphere of North American pollution to the North Atlantic and on to Europe, as will be discussed below. Another pathway for transatlantic transport of North American pollution involves strong convection over the central and eastern United States, particularly in summer, that lifts ozone and its precursors to the middle and upper troposphere



**Figure 7.** (top) Cumulative probability distributions and (bottom) scatterplots of measured and simulated surface ozone and CO for 1994–1997 at Mace Head (1994–1996, light dots; 1997, dark dots). The  $y = x$  lines are also shown. Data points are 24-hour averages. The cumulative probability distribution is plotted on a probability scale such that a normal distribution would plot as a straight line.

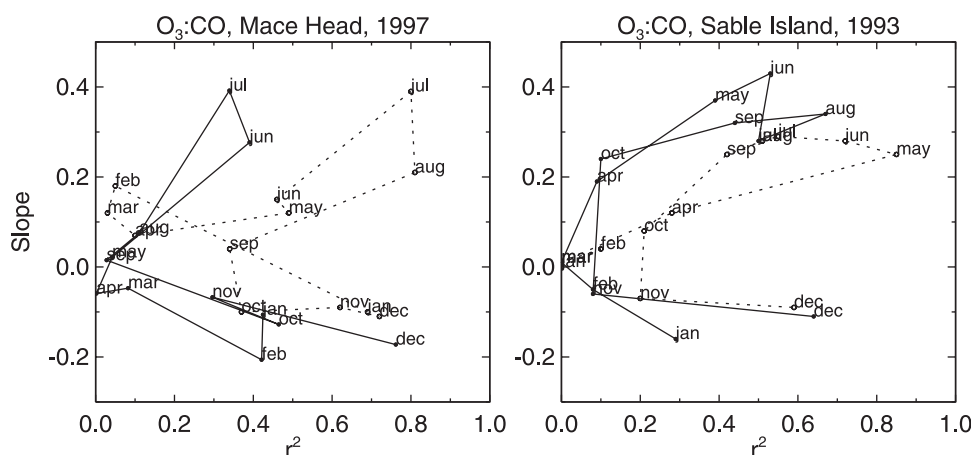
where they are exported by the westerlies [Jacob *et al.*, 1993a; Thompson *et al.*, 1994]. Yet another mechanism for transatlantic transport of North American pollution involves frontal lifting over the western North Atlantic by WCBs [Stohl, 2001; Stohl *et al.*, 2002].

[15] The prevailing westerlies at northern midlatitudes generally preclude direct transatlantic transport of European pollution to North America, although we will see that such conditions can occasionally occur. A more general route for transatlantic transport of European pollution involves circulation around the southern branch of the Bermuda-Azores high, particularly in summer, when the high extends far to the north and east (Figure 2c). Evidence for such transport of European pollution has been seen in aerosol observations in the Canary Islands [Raes *et al.*, 1997] and at Barbados

[Hamelin *et al.*, 1989; Li-Jones and Prospero, 1998]. For ozone, as we will see, the short lifetime at low latitudes limits the efficiency of this transport. European pollution can also be transported to North America by circulation around the Icelandic low, and by westerly transport around the northern midlatitude band. We will quantify the importance of these different processes.

[16] Figure 3 shows the simulated monthly average transport fluxes (0–3-km altitude) and surface air concentrations of North American anthropogenic CO and North American ozone in January, April, July, and October 1997. Figure 4 shows the same for European anthropogenic CO and European ozone. “Surface air,” here and in what follows, refers to the lowest model layer (0–100 m above the surface). The model does not resolve the nighttime depletion of surface





**Figure 8.** Observed (solid lines) and simulated (dashed lines)  $O_3:CO$  correlations (left) at Mace Head for 1997 and (right) at Sable Island for 1993. The slope of the regression line is plotted against the square of the correlation coefficient for individual months.

ozone often seen at continental sites because of loss by deposition and chemistry in a shallow surface layer (a few tens of meters deep). As a result, model results are expected to be representative of surface ozone concentrations over continents only in the daytime, when the mixed layer is sufficiently deep to be resolved by the model [Jacob *et al.*, 1993b]. This is not an issue for the marine sites used for model evaluation in section 4.

[17] We see from Figure 3 that surface concentrations of North American anthropogenic CO are highest in winter and lowest in summer, due to more efficient boundary layer ventilation and chemical loss of CO in summer. Export fluxes of North American anthropogenic CO in the lower troposphere are correspondingly strongest in winter and weakest in summer (left panels, Figure 3). Surface concentrations of North American ozone in contrast are highest in summer and lowest in winter, due to more active photochemical ozone production in summer. Export fluxes of North American ozone are strongest in spring and summer (right panels, Figure 3).

[18] We find that transatlantic transport of North American ozone and CO takes place in the lower troposphere year round, and transport in the middle and upper troposphere is also important in summer. Compared to transpacific transport of Asian pollution to North America, which takes place mainly in the free troposphere followed by subsidence [Jacob *et al.*, 1999; Yienger *et al.*, 2000; Fiore *et al.*, 2002], the relatively short distance between North America and Europe and the prevailing westerly flow extending down to the surface favor transport in the boundary layer. Over eastern Asia, in contrast, westerly winds do not extend down to the surface, so that lifting to the free troposphere is necessary to enable transpacific transport [Bey *et al.*, 2001b].

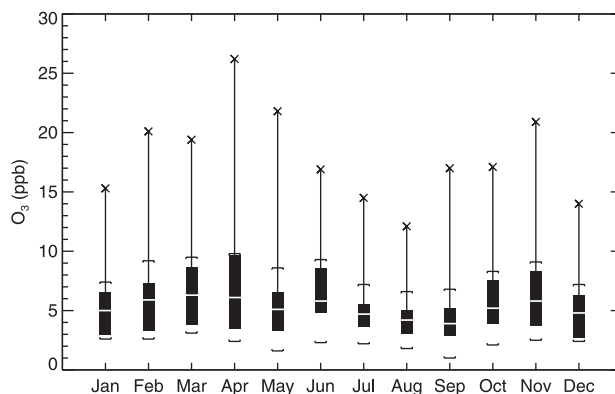
[19] Figure 4 illustrates the mean transatlantic flow patterns for European anthropogenic CO and European ozone. Transport of pollution from Europe to North America in winter is mainly by the general westerly circulation. In summer, that circulation is considerably weaker, and European influence on North America takes place principally by transatlantic circulation around the

Bermuda-Azores high and the Icelandic low. The influence of European sources on surface ozone in North America is minimum in summer due to the weak circulation and the short lifetime of ozone.

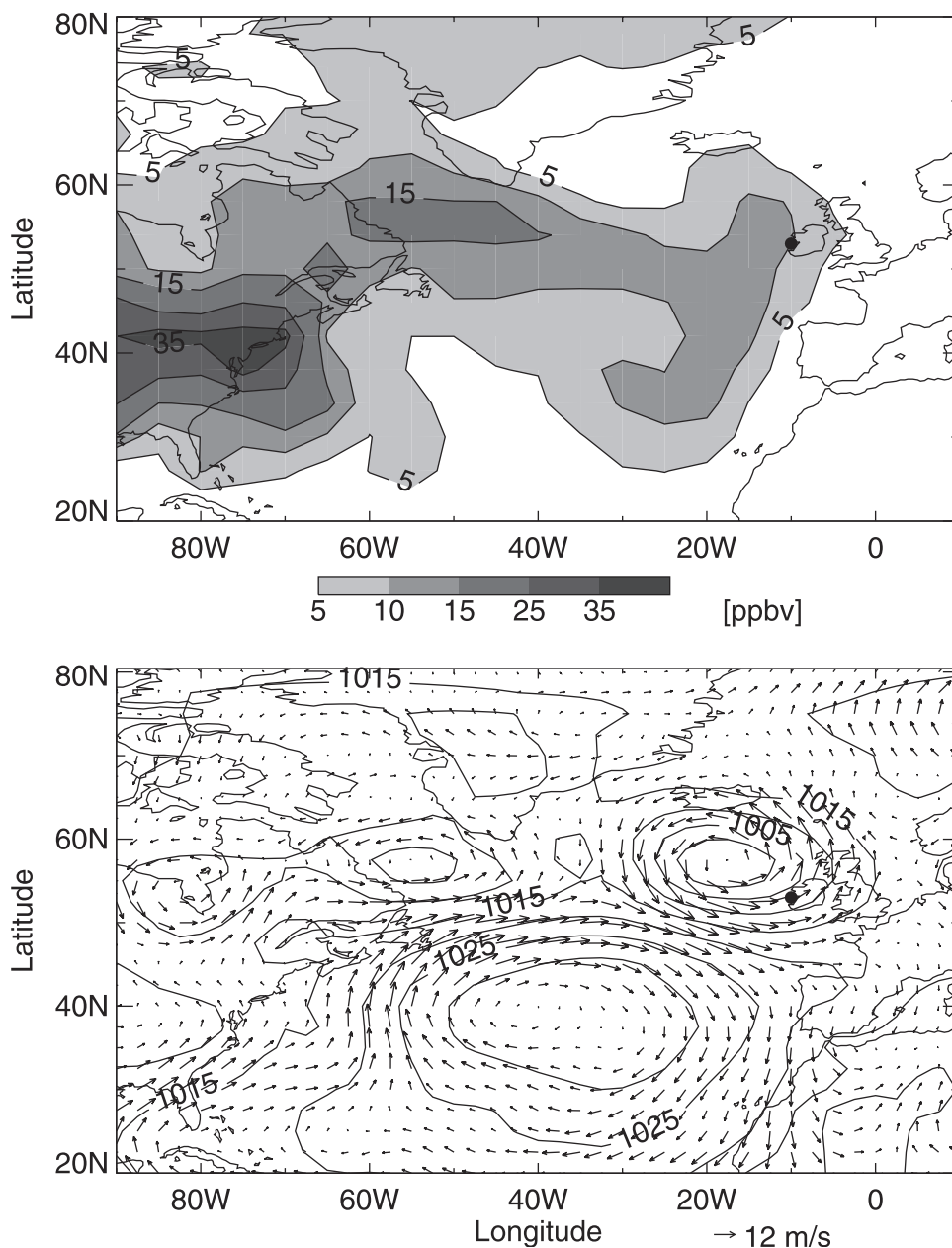
## 4. Time Series of Ozone and CO at North Atlantic Sites

### 4.1. Mace Head

[20] Ozone and CO measurements at Mace Head are available for 1994–1997. Time series for 1997 are shown in Figures 5 and 6. The model results are superimposed. The simulated contributions from North American ozone and North American anthropogenic CO are also shown using tagged tracers. The simulated contributions from other source regions are shown and discussed in a separate report [Li *et al.*, 2001b]. Statistics of comparisons between the



**Figure 9.** Simulated seasonal variation of the North American contribution to surface ozone at Mace Head. Values are 5-year statistics (1993–1997) of 24-hour average data. Shown are monthly averages (white bars) and standard deviations (brackets). The black bars indicate the inter-annual range of the monthly averages. The whiskers represent the monthly maxima over the 5-year period.



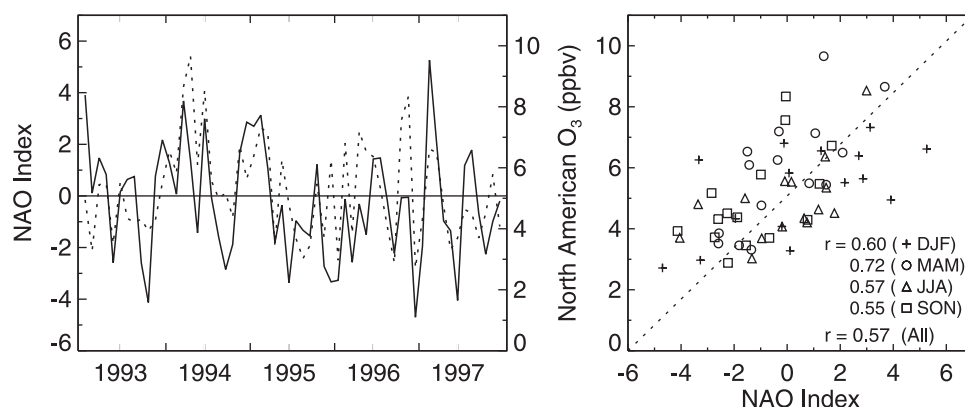
**Figure 10.** North American pollution episode at Mace Head: simulated daily average (top) surface concentrations of North American ozone on 18 June 1997 and (bottom) GEOS sea level pressure and 925-hPa winds on the same day. The solid circle indicates the location of Mace Head.

observed and simulated time series for 1994–1997 are shown in Figure 7. For ozone the model captures the background concentrations, the probability distributions, the frequencies and magnitudes of pollution events, and the day-to-day variations. Similarly, good agreement is found for CO. The simulated mean concentrations of ozone and CO for 1994–1997 are 34 ppbv and 137 ppbv, respectively, in good agreement with observed means (34 ppbv and 146 ppbv). The correlation coefficients between the model results and measurements are 0.62 for ozone and 0.71 for CO.

[21] Figure 8 (left panel) shows the observed and simulated  $O_3$ :CO correlations at Mace Head for 1997. Significant positive correlations are found in the summer months;

correlation coefficients are higher in the model, as would be expected since the model has fewer factors of variability than the observations. The slope of the  $O_3$ :CO relationship in the model in summer ( $0.3 \text{ mol mol}^{-1}$ ) is consistent with observations and tests the simulation of photochemical ozone production in a manner relatively independent of model transport [Chin *et al.*, 1994; Atherton *et al.*, 1996]. Ozone and CO are anticorrelated in the winter, both in the model and in the observations, due to titration by  $NO_x$  emissions [Derwent *et al.*, 1994; Parrish *et al.*, 1998].

[22] We find that all major high- $O_3$  and high-CO events at Mace Head are of European origin in our 5-year simulation [Li *et al.*, 2001b]. North American ozone contributes only 5 ppbv on average (Figure 5), while North American anthro-



**Figure 11.** (left) Time series and (right) scatterplots of the monthly average North Atlantic Oscillation (NAO) index (solid line) and simulated North American ozone at Mace Head (dashed line) for 1993–1997. The NAO index is defined as the difference of normalized sea level pressure between Reykjavik (Iceland) and Gibraltar (United Kingdom) [Jones *et al.*, 1997] and was obtained from the Climatic Research Unit, University of East Anglia, at <http://www.cru.uea.ac.uk/cru/data/nao.htm>.

pogenic CO contributes on average 10–20 ppbv (Figure 6). Asian ozone influence is negligible (never more than 2 ppbv) over the 1993–1997 period. The 5-year monthly statistics of the simulated North American contribution to surface ozone at Mace Head are shown in Figure 9. There is relatively little seasonal variation in the monthly means, and about a factor of 2 interannual variability. The largest interannual variability and the largest events are in spring and fall, reflecting a combination of efficient ozone production, long ozone lifetime, and fast transport [Wang *et al.*, 1998b].

[23] During transatlantic transport events in the model, North American ozone contributes up to 10–15 ppbv in summer and 15–25 ppbv in other seasons to surface ozone at Mace Head, while North American anthropogenic sources contribute up to 40 ppbv CO. For purpose of later discussion, we define a transatlantic transport event as a North American contribution to surface ozone at Mace Head of 10 ppbv or more. The events for 1997 are highlighted with arrows in Figures 5 and 6. We find that these transatlantic transport events occur in every season of the year in the 5-year simulation (Figure 9).

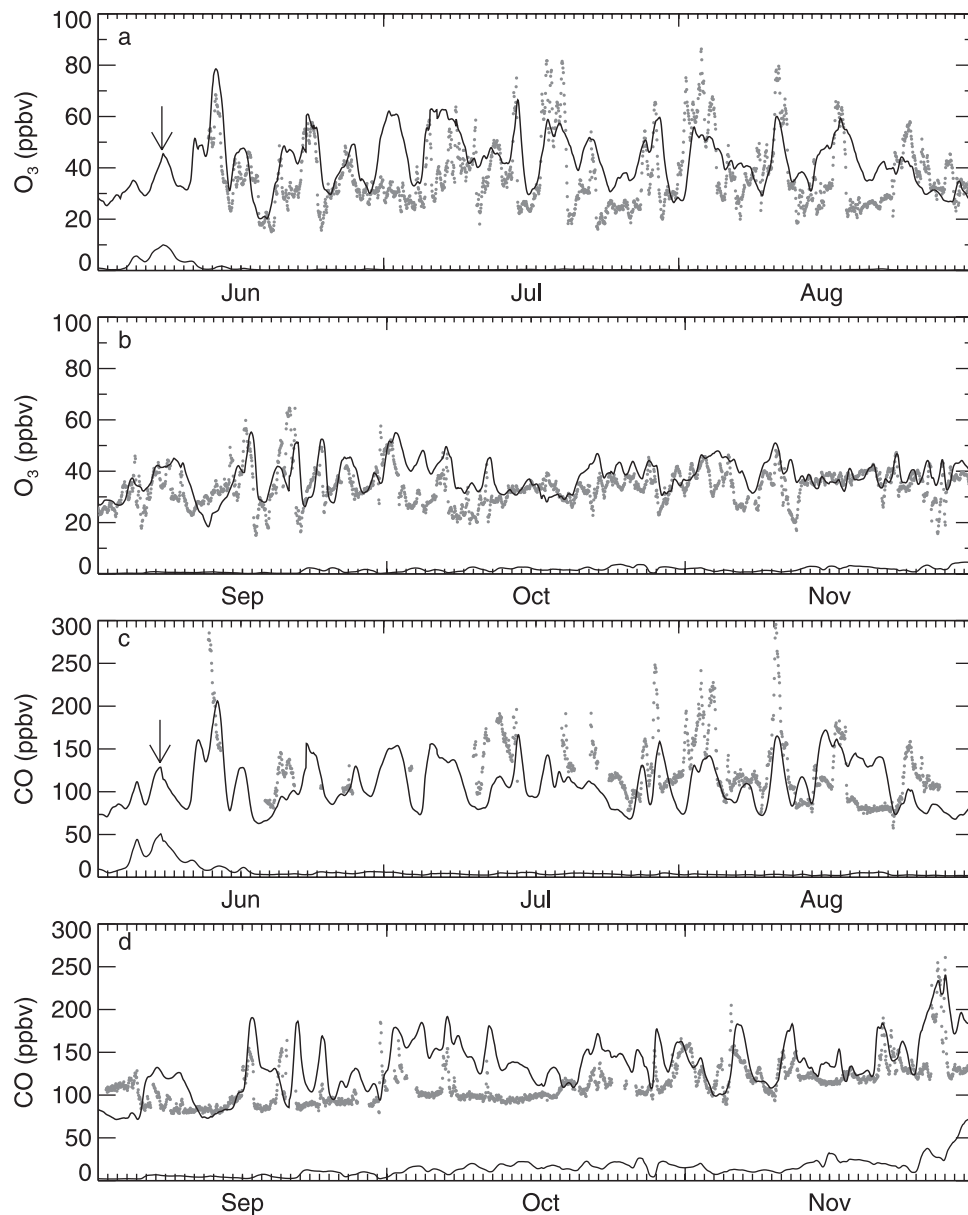
[24] A question to be addressed is whether the transatlantic transport events seen in the model would be detectable in the observations. We find that these events correspond to secondary peaks in the observations of ozone and CO, for example, on 18 June, 25 July, 1 August, and 21 August (Figures 5 and 6). The strongest event in our 5-year simulation (26 ppbv) is on 10 April 1994. Examination of the observations reveals an increase from 40 ppbv before 10 April to 50 ppbv on 10 April. Observed CO concentrations similarly increased from 180 ppbv before 10 April to 260 ppbv on 10 April. Two transatlantic transport events in March and November 1994 previously identified by Ryall *et al.* [1998] from their particle dispersion model analysis of CFCs at Mace Head are also seen in our 1994 tagged ozone and CO simulations.

[25] We examined the meteorological pattern for each of the transatlantic transport events identified by the model simulation for 1993–1997. Figure 10 illustrates a typical case (18 June 1997). An intense Icelandic low between Iceland and the British Isles channeled outflow from North America to Mace Head. In contrast, the mean summer flow

patterns in Figures 2 and 3 indicate that the North American outflow over the eastern North Atlantic diverges to the Arctic and to the tropics, largely missing Europe. Transport of North American pollution to Mace Head on 18 June and during other episodes is largely confined to below 3-km altitude in the model, consistent with air back-trajectories (M. Evans, Harvard University, personal communication, 2001). This particular event lasted until 20 June, when the low-pressure system moved eastward, channeling the flow further to continental Europe. As will be discussed in section 5, transatlantic transport events in the model at Mace Head are forerunners of events over continental Europe. Such transatlantic transport in the lower troposphere is also seen in the analysis of intercontinental transport pathways by Stohl *et al.* [2002] using a Lagrangian particle dispersion model. For the entire 1993–1997 record, we find that the strength and location of the Icelandic low can be used as an indicator of the occurrence of transatlantic transport events at Mace Head and in Europe.

[26] Long-term meteorological observations have shown that a stronger than normal Icelandic low is usually accompanied by a stronger than normal Bermuda-Azores high, a phenomenon called the North Atlantic Oscillation (NAO) [Walker and Bliss, 1932; Van Loon and Rogers, 1978]. The NAO is recognized as a major factor of interannual variability in temperature and precipitation over Europe [Hurrell, 1995]. It is measured by the NAO index, representing the difference of normalized sea level pressure between a station in Iceland and one in the Azores [Rogers, 1984, 1997], Lisbon (Portugal) [Hurrell, 1995; Hurrell and Van Loon, 1997], or Gibraltar (United Kingdom) [Jones *et al.*, 1997]. Sea level pressures are normalized by dividing monthly anomalies (compared to the long-term mean) by long-term monthly standard deviations. The positive phase of the NAO is characterized by a strong Icelandic low and Bermuda-Azores high and hence a strong north-south pressure gradient and, as a result, strong surface westerlies across the North Atlantic onto Europe. The pressure gradient is weak in the negative phase of the NAO.

[27] The left panel of Figure 11 shows a time series of the monthly mean NAO index together with our simulated

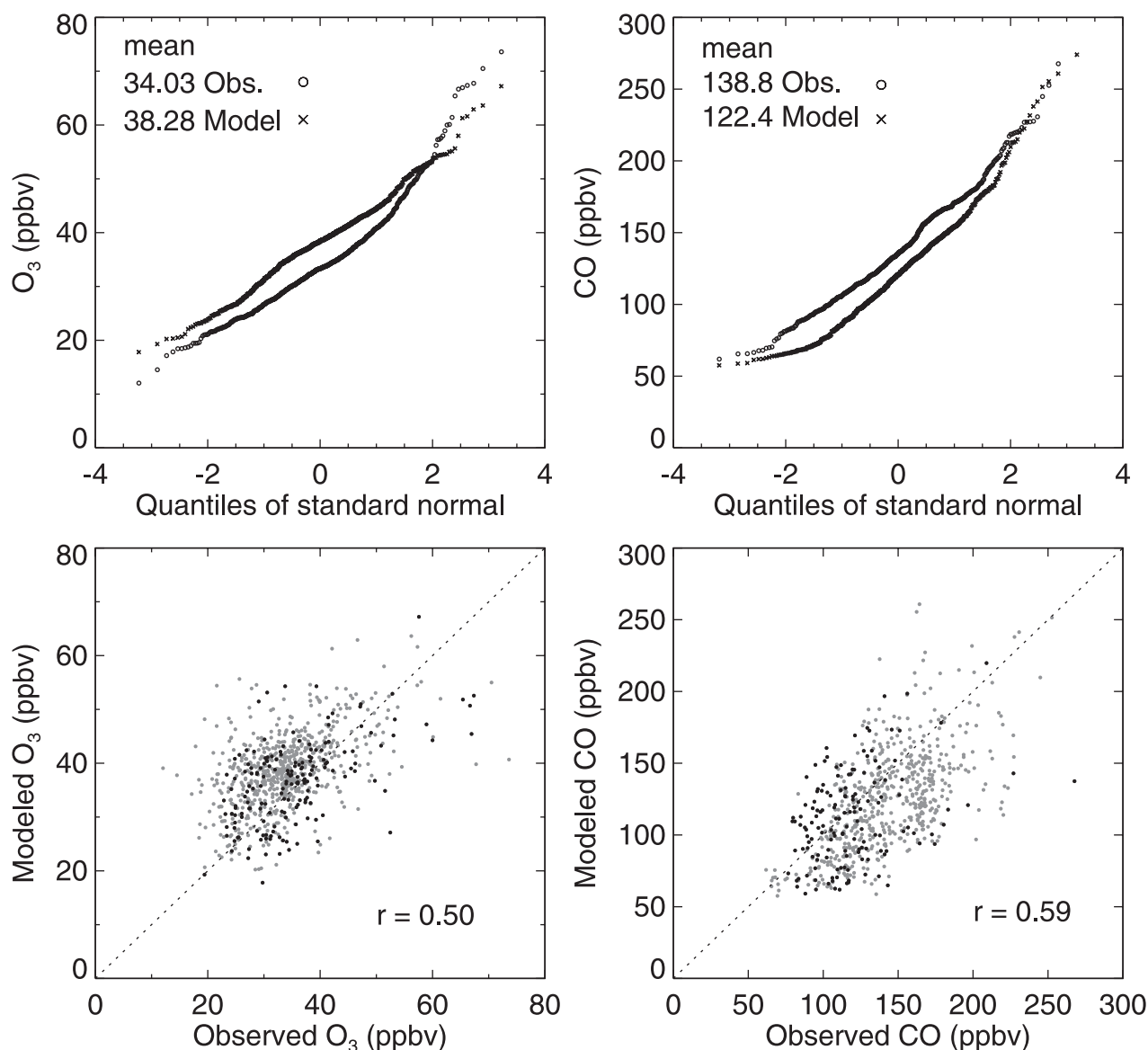


**Figure 12.** Hourly average surface concentrations of (a and b) ozone and (c and d) CO for June–November 1997 at Sable Island (44°N, 60°W) (gray dots, observations; thick lines, model). Contributions from European ozone and CO in the model are shown as thin lines. The arrows indicate a major transatlantic transport event.

monthly mean North American ozone at Mace Head for 1993–1997. The right panel of Figure 11 shows the correlation between these two quantities over that record. There is a strong correlation ( $r = 0.57$ ), particularly in spring ( $r = 0.72$ ). The NAO index used here is from Jones *et al.* [1997], i.e., based on pressure difference between Reykjavik (Iceland) and Gibraltar. We find that NAO indices based on other definitions (see references above) show similar correlations. The high correlation implies that the NAO index can be used as a predictor for the transatlantic transport of North American pollution to Mace Head and on to Europe.

[28] Recent studies have shown that the NAO exhibits coherent variability on decadal timescales, both in observations [Hurrell, 1995; Jones *et al.*, 1997; Sutton and Allen,

1997; Rajagopalan *et al.*, 1998] and in coupled ocean-atmosphere general circulation models (GCMs) [Delworth and Mehta, 1998; Grötzner *et al.*, 1998; Rodwell *et al.*, 1999; Mehta *et al.*, 2000]. There is also considerable month-to-month variability, as is apparent in Figure 11. Most studies of the interannual variability of the NAO have focused on December–March, when the NAO exerts strong influence on the climate of the Northern Hemisphere and the index exhibits the strongest interdecadal variability. The December–March 1993–1997 period was in a strong positive phase of the NAO, with the exception of 1996 which shows a negative NAO index [Hurrell, 1995; Jones *et al.*, 1997], but the monthly data show a range of positive and negative values for each year (Figure 11). Using GCM with anthropogenic



**Figure 13.** (top) Cumulative probability distributions and (bottom) scatterplots of simulated surface ozone and CO versus measurements for 1993, 1994, and 1997 at Sable Island (1993 and 1994, light dots; 1997, dark dots). The  $y = x$  lines are also shown. Data points are 24-hour averages. The cumulative probability distribution is plotted on a probability scale such that a normal distribution would plot as a straight line.

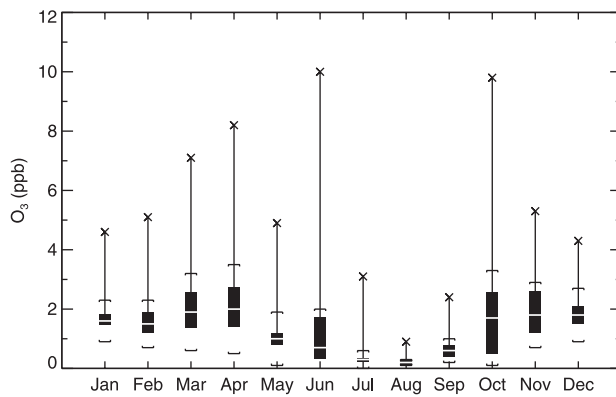
forcing from greenhouse gases and sulphate aerosol, *Osborn et al.* [1999] predicted a significant decline in the NAO index over the 21st century. Such a decline would reduce the transatlantic transport of North American pollution to Europe. On a year-to-year timescale, the predictability of the NAO in GCMs [*Sutton and Allen, 1997; Griffies and Bryan, 1997; Rodwell et al., 1999; Mehta et al., 2000; Bretherton and Battisti, 2000*] suggests that one could in this manner forecast not only precipitation and temperature in Europe, but also the transatlantic transport of pollution.

#### 4.2. Sable Island

[29] Ozone and CO measurements at Sable Island are available for 1993, 1994, and June–November 1997. Previously, *Lin et al.* [1998] used a 3-D regional chemistry

model to simulate time series of ozone and CO observed in August 1993 at the site. Time series for June–November 1997 are shown in Figure 12. Model results are superimposed; the European contributions to ozone and CO, which are in general small (less than 2 ppbv for ozone), are also shown using tagged tracers. Contributions from other source regions are presented by *Li et al.* [2001b]. North American sources are dominant by far. Asian influence on ozone is less than 5 ppbv anytime. One major European pollution event at Sable Island is found in early June, which is highlighted by arrows in Figure 12 and will be discussed below.

[30] Statistics of comparisons between the observed and simulated time series for the ensemble of 1993–1997 observations are shown in Figure 13. For ozone, the model has a



**Figure 14.** Simulated seasonal variation of the European contribution to surface ozone at Sable Island. Values are 5-year statistics of 24-hour average data (1993–1997). Shown are monthly averages (white bars) and standard deviations (brackets). The black bars indicate the interannual range of the monthly averages. The whiskers represent the monthly maxima over the 5-year period.

positive bias of 4 ppbv but otherwise has some success in capturing the probability distributions, the frequencies and magnitudes of pollution events, and the day-to-day variations. Similar positive bias in the GEOS-CHEM simulated surface ozone in the United States has been found by *Fiore et al.* [2002]. There is a negative bias of 15 ppbv in the simulated CO concentrations. The simulated mean concentrations of ozone and CO are 38 ppbv and 122 ppbv, respectively, while the observed means are 34 ppbv and 139 ppbv. The correlation coefficients between the model results and measurements are 0.50 for ozone and 0.59 for CO.

[31] Shown in the right panel of Figure 8 are the observed and simulated  $O_3:CO$  correlations at Sable Island for 1993, when a full year of observations is available. The magnitude and seasonal variation of the  $O_3:CO$  slope are similar to Mace Head, both in the model and in the observations. The ozone pollution season (positive  $O_3:CO$  slope) at Sable Island extends longer than that at Mace Head, reflecting the higher latitude of Mace Head and the generally cooler and cloudier weather in Europe [*Doddridge et al.*, 1994].

[32] Statistics of European influence on surface ozone at Sable Island during the 5-year simulation are shown in Figure 14. Monthly average European contributions are less than 2 ppbv throughout the year, but there are occasional events of 5–10 ppbv which occur in all 5 years. Similar to Mace Head, the transatlantic transport events at Sable Island occur mostly in spring and fall. The 1997 simulation indicates a particularly intense summertime event of transatlantic transport from Europe to Sable Island on 3–8 June 1997 (Figure 12). During the event, about 10 ppbv of ozone and 30–50 ppbv of CO are of European origin. Unfortunately, there are no measurements for that time period. This event at Sable Island was related to the event seen several days earlier on 29 May to 1 June at Mace Head, with observed peak concentrations of 70 ppbv ozone and 200 ppbv CO (Figures 5 and 6).

[33] Figure 15 illustrates the flow driving the European pollution event on 3–8 June 1997 at Sable Island. On 3 June, two low-pressure systems were merging to form an

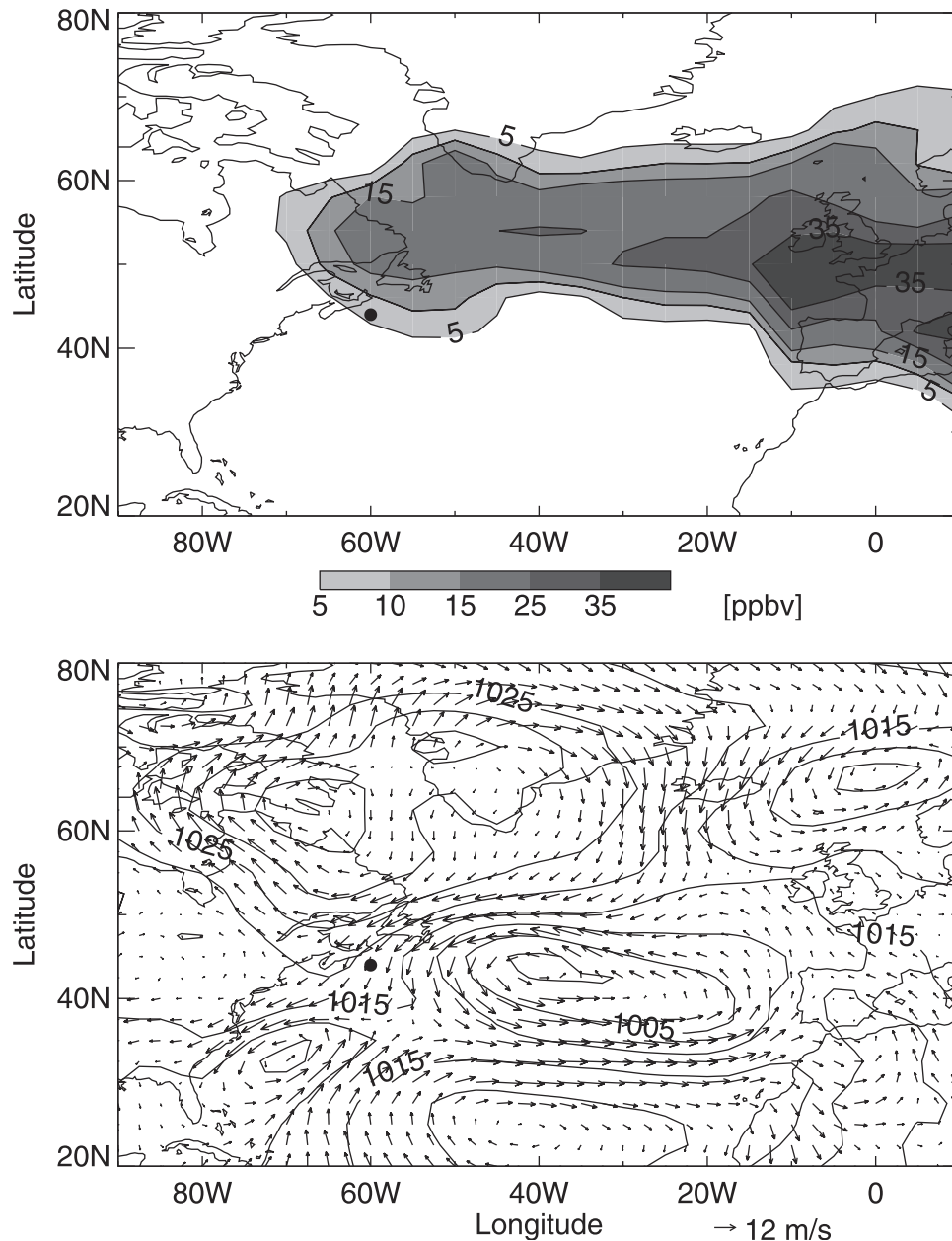
extensive low-pressure system along 45°N latitude across the North Atlantic, while an extensive high-pressure system was centered over Labrador. Thus an easterly flow from Europe to North America was established below 3-km altitude. This easterly flow is highly anomalous compared to the mean westerly flow pattern over the region (Figures 2 and 4). It eventually broke up on 7 June, when the low-pressure system moved northeastward to Europe. The European ozone pollution persisted over northeast North America until 11 June, when it extended as far south as Bermuda (5 ppbv European contribution to surface ozone).

[34] We examined the other major European pollution events at Sable Island during the 5-year simulation and found they are all associated with similar anomalous direct transatlantic transport in the boundary layer. The more typical mechanisms for transport of European pollution to North America including northern circulation around the Icelandic low, southern circulation around the Bermuda-Azores high, and transport around the globe in the westerlies (see section 3) never generate high- $O_3$  events over North America.

## 5. Surface Ozone Enhancements From Intercontinental Transport of Pollution

[35] Figure 16 shows the enhancements in the mean surface ozone concentrations in June–August 1997 over the Northern Hemisphere due to anthropogenic emissions from North America, Europe, and Asia, as determined from sensitivity simulations with these emissions shut off. These enhancements add up together to much less than the ozone concentrations in the standard simulation, because of contributions from natural sources as well as chemical non-linearity. Shutting off North American anthropogenic emissions decreases the mean surface ozone concentrations in Europe by 2–4 ppbv, with maximum effect in northwest Europe. The relatively large North American influence over northern Africa and the Middle East is due to the deep boundary layer mixing over these arid regions which entrains North American ozone from aloft [*Li et al.*, 2001a].

[36] We find that long-range transport of ozone itself is far more important in contributing to North American influence on surface ozone in Europe than transport of the precursors ( $NO_x$  and PAN) followed by ozone production in the European boundary layer. By difference with the standard simulation, we find that the monthly average influx of North American pollution ozone to the European boundary layer is 1.4 Gmol  $d^{-1}$  in July 1997, while ozone production in the European boundary layer resulting from the influx of North American ozone precursors is 0.2 Gmol  $d^{-1}$ . Similarly, *Jacob et al.* [1999] found that Asian influence on surface ozone in the United States is driven by long-range transport of ozone rather than of its precursors. This influx of North American pollution ozone to the European boundary layer can be compared to its export out of the North American boundary layer. Our model simulation shows a monthly mean export flux of pollution ozone from the North American boundary layer of 6 Gmol  $d^{-1}$  in July 1997, out of a net production in that region of 8.3 Gmol  $d^{-1}$ , and this export is mostly by convection over the continent [*Jacob et al.*, 1993a]. The eastward flux of North



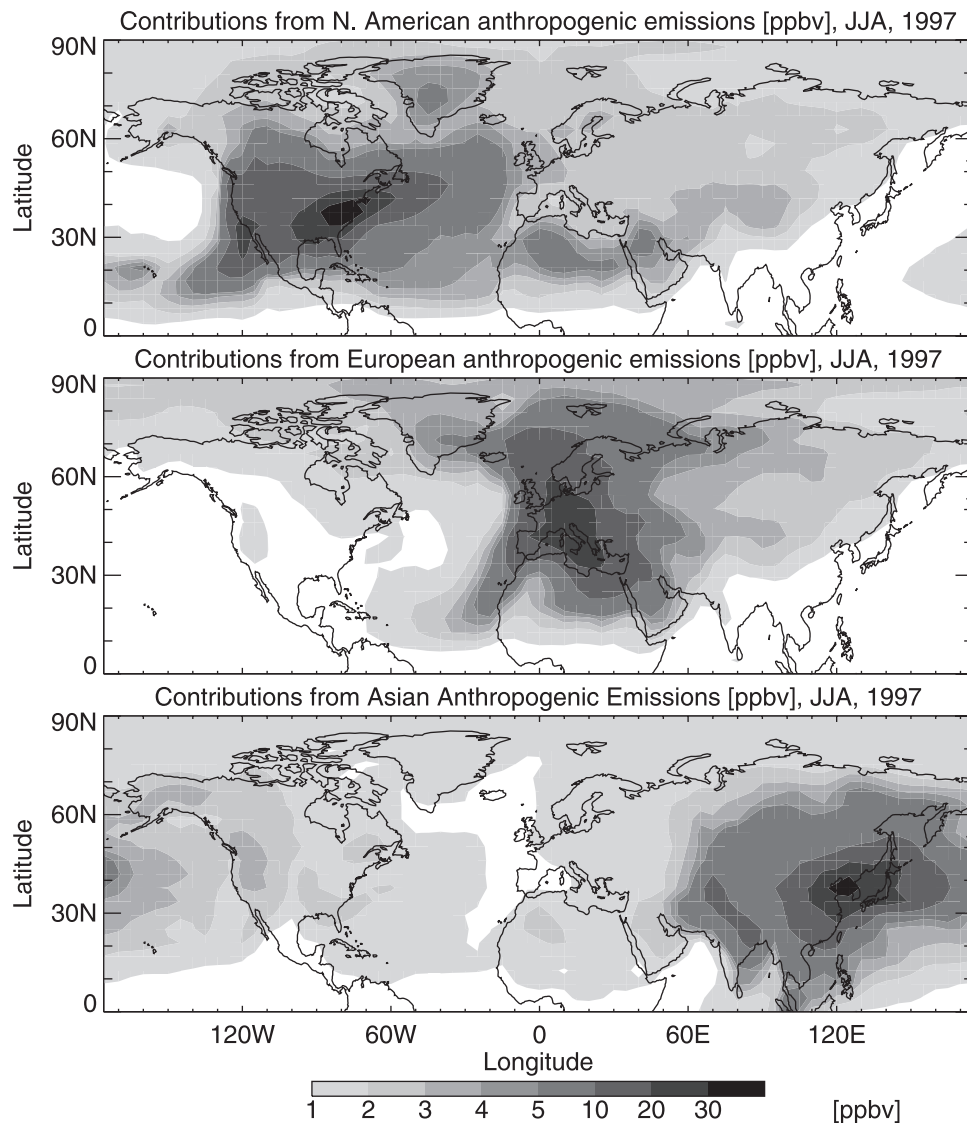
**Figure 15.** European pollution episode at Sable Island: simulated daily average (top) surface concentrations of European ozone on 3 June 1997 and (bottom) GEOS sea level pressure and 925-hPa winds on that day. The solid circle indicates the location of Sable Island.

American pollution ozone across the western North Atlantic ( $60^{\circ}\text{W}$ ) is  $10 \text{ Gmol d}^{-1}$ , higher than the export from the North American boundary layer because of subsequent production from exported ozone precursors. These results are consistent with previous 3-D model studies of the export of ozone pollution from North America [Chin *et al.*, 1994; Liang *et al.*, 1998] and imply that only 10–20% of this export goes on to enter the European boundary layer.

[37] We examined the relative contributions of direct boundary layer advection versus subsidence from the free troposphere in contributing to North American influence on surface ozone in Europe in July 1997, and find the two to be of comparable importance. The mean influx of North American ozone to Europe from transport in the boundary

layer below 2.6-km altitude is  $0.8 \text{ Gmol d}^{-1}$  in the model, while downward influx from subsidence across the 2.6-km surface over Europe is  $0.6 \text{ Gmol d}^{-1}$ . Again, these results were obtained by difference between the standard simulation and the simulation with North American anthropogenic emissions shut off.

[38] The tagged tracer analysis in section 4 identified several events of transatlantic transport from North America to Mace Head during the summer of 1997, for instance, on 18–20 June and 14–16 July (Figures 5 and 6). The sensitivity simulation with North American anthropogenic emissions shut off identifies the same events with comparable magnitudes. We find that these events are the forerunners of more general North American pollution events



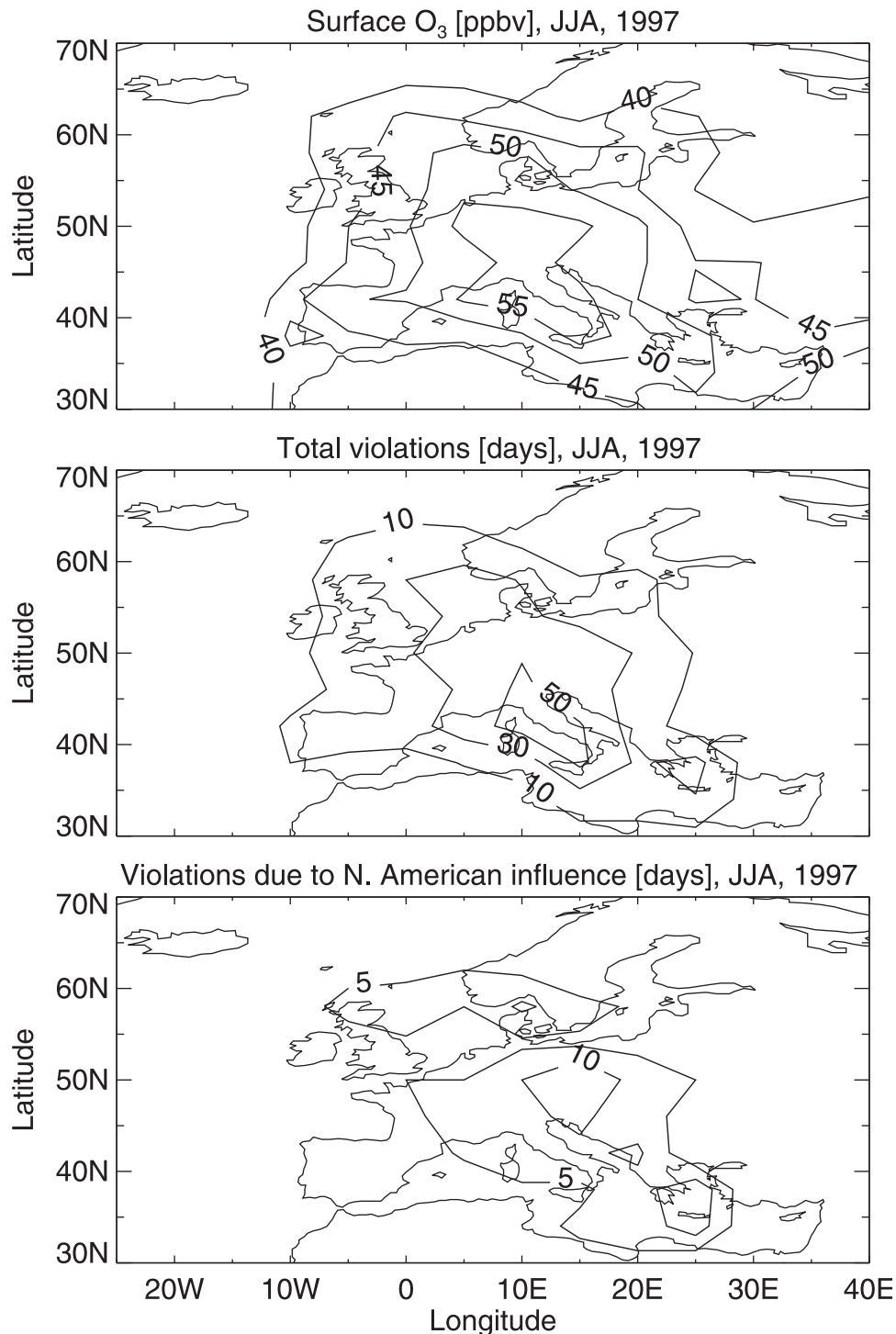
**Figure 16.** Sensitivity of simulated surface ozone concentrations in June–August 1997 to anthropogenic emissions in the individual continents. The plots show the mean decreases in concentrations when anthropogenic emissions from (top) North America, (middle) Europe, and (bottom) Asia are shut off separately. “Surface ozone” refers to the lower mixed layer (0–100 m), i.e., the lowest layer of the model. See color version of this figure at back of this issue.

over the European continent during which North American anthropogenic emissions enhance surface ozone in the British Isles by up to 10 ppbv and in continental Europe by up to 8 ppbv (daily mean). For example, the North American plume observed on 18–20 June at Mace Head moved across the European continent, enhancing surface ozone levels by 4–8 ppbv in western Europe on 21 June and by 4–6 ppbv in central and eastern Europe on 23 June (not shown).

[39] Such North American influence on surface ozone in Europe has important implications for achieving European ozone air quality standards. The current European Council Directive on air pollution by ozone (Council Directive 92/72/EEC) defines a health-based threshold of 55 ppbv (8-hour average) and a vegetation protection threshold of 65 ppbv (24-hour average) [European Council of Ministers,

1992]. The World Health Organization (WHO) has recommended an air quality guideline of 60 ppbv (8-hour average) for Europe [World Health Organization, 1996]. Figure 17 (top panel) shows the mean simulated surface ozone concentrations in Europe in July 1997. Maximum concentrations are in the southeast, consistent with observations [Scheel *et al.*, 1997; Kley *et al.*, 1997]. As pointed out in section 3, “surface” concentrations in the model are actually for the lower mixed layer (0–100-m altitude), and the corresponding 24-hour averages are more representative of 8-hour daily maxima rather than of 24-hour averages in surface air. With this assumption, Figure 17 (middle panel) shows the number of daily violations of the 55 ppbv ozone standard in the model out of 92 days for June–August 1997. Violations are widespread, exceeding half of summer days over much of Italy. Figure 17 (bottom

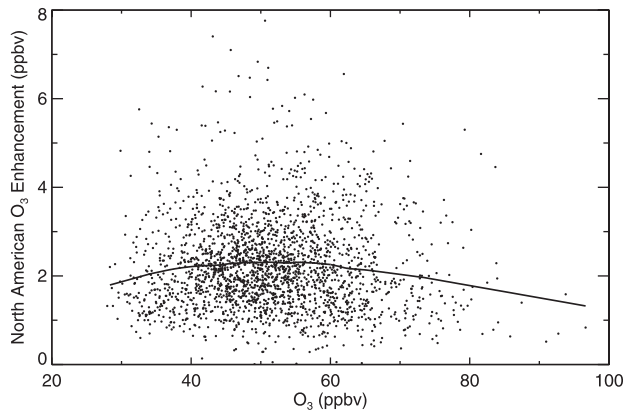




**Figure 17.** (top) Simulated mean surface ozone concentrations for June–August 1997 in Europe; “surface” actually refers to the lower mixed layer (0–100-m altitude), i.e., the lowest layer of the model. (middle) The number of days (out of 92) when the ozone standard of 55 ppbv is exceeded in the simulation. (bottom) The number of these days when the ozone standard would not have been exceeded in the absence of North American influence.

panel) shows the number of violations that would not occur in the absence of North American anthropogenic emissions. There are 5–10 such violations over much of Europe for the summer of 1997, with the largest number (>10) in central Europe. The number of violations obviously depends on the

formulation of the ozone standard, but we find that the pattern of North American influence is similar for different ozone standards. For example, we find that for higher standards of 65 ppbv and 75 ppbv, there are 2–5 and less than 2 violations, respectively, that would not occur in the



**Figure 18.** Simulated surface ozone enhancement over continental Europe from North American pollution, as determined by difference between the standard simulation and a simulation with North American anthropogenic emissions shut off, plotted as a function of local surface ozone concentrations. Values are daily (24-hour) averages for June–August 1997 and for the ensemble of continental Europe grid squares in the model. The curve is a locally weighted regression.

absence of North American anthropogenic emissions over much of Europe for the summer of 1997.

[40] Ozone concentrations in the range 50–60 ppbv, at the threshold of the European standards, are, in fact, particularly susceptible to intercontinental transport of pollution. This was recently shown by *Fiore et al.* [2002] for the United States. It is shown here for continental Europe in Figure 18 as the ozone enhancement from North American anthropogenic emissions plotted as a function of the daily local surface ozone concentration, for the ensemble of continental Europe grid squares in June–August 1997. High ozone concentrations are usually associated with regional stagnation episodes when transport from outside Europe is suppressed. Low ozone concentrations tend to be associated with tropical rather than North American influence.

[41] Shutting off European anthropogenic emissions in the model leads to a decrease of less than 2 ppbv in the monthly average surface ozone in North America in summer (middle panel, Figure 16). This again is consistent with our results using tagged ozone tracers for Sable Island in section 4. European pollution influence on surface ozone over eastern Asia is also weak, less than 2 ppbv on average, because of the prevailing Asian monsoon; greater European influence on Asia is found in spring (Liu et al., submitted manuscript, 2001).

[42] Shutting off Asian anthropogenic emissions in the model leads to a decrease of 1–4 ppbv in the monthly average ozone concentrations at the surface in North America in June–August 1997, with the maximum influence in the western United States (bottom panel, Figure 16). This Asian influence in North America is examined in more detail by *Fiore et al.* [2002]. We find that Asian pollution contributes less than 2 ppbv to surface ozone over Europe in the summer of 1997 (bottom panel, Figure 16).

[43] Figure 19 shows the higher-altitude enhancements in the mean ozone concentrations at 45°N latitude in June–

August 1997 due to anthropogenic emissions from North America, Europe, and Asia as determined from sensitivity simulations with these emissions shut off. We see that North American anthropogenic emissions enhance ozone in the middle-upper troposphere over Europe by about 10 ppbv on average. The impact of European anthropogenic emissions is largely limited to the lower troposphere, reflecting the relatively weak convective and frontal activity over Europe as compared to North America or Asia [Stohl, 2001a; Stohl et al., 2002].

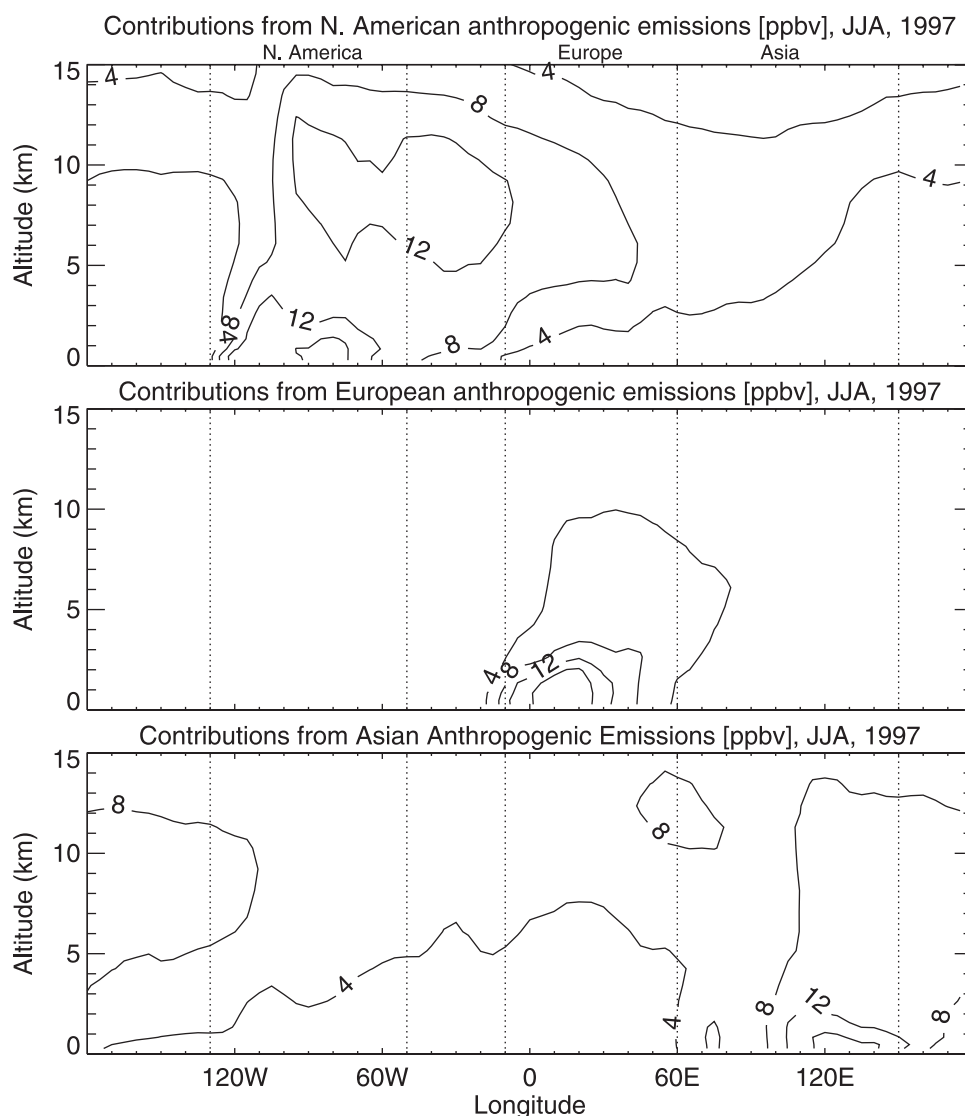
## 6. Summary and Conclusions

[44] We have used a 5-year (1993–1997) simulation of tropospheric ozone- $\text{NO}_x$ -hydrocarbon chemistry with the GEOS-CHEM global 3-D model to quantify the impact of transatlantic transport of pollution on surface ozone in Europe and North America. Measurements of ozone and CO at Mace Head (Ireland) and Sable Island (Canada) were used to evaluate transatlantic transport in the model. The model reproduces well the frequencies and magnitudes of pollution events at these sites. The contributions from transatlantic transport of pollution are examined in the model through a combination of sensitivity simulations and tagged tracer analyses.

[45] The contribution from North American pollution to surface ozone at Mace Head is 5 ppbv on average, with little seasonal variability, and up to 10–20 ppbv during transatlantic transport events. We find that these events are associated with low-level westerly flow (below 3-km altitude) channeled to northwest Europe by a strong Icelandic low centered between Iceland and the British Isles. We further find that transatlantic transport events at Mace Head are forerunners of broader events of long-range transport of North American pollution to the surface of Europe. North American ozone influence at Mace Head and more generally over Europe is strongly correlated with the North Atlantic Oscillation (NAO) index. This result suggests that long-term forecasts of the NAO, which are viewed with considerable interest for predicting precipitation and temperature in Europe, could also serve to predict the transatlantic transport of pollution.

[46] European pollution influence on surface ozone at Sable Island is less than 2 ppbv on average and up to 5–10 ppbv during transatlantic transport events. These events are associated with low-level easterly flow (below 3-km altitude) driven by anomalous low-pressure systems at 45°N over the North Atlantic. More typical pathways for transport of European pollution to North America include the circulations around the northern branch of the Icelandic low and around the southern branch of the Bermuda-Azores high, as well as the westerly flow around the globe. However, we find that none of these pathways lead to events of high European ozone over North America in our 5-year simulation record.

[47] Our sensitivity simulation for the summer of 1997 indicates that North American anthropogenic emissions enhance surface ozone in Europe by 2–4 ppbv on average, and by 5–10 ppbv during transatlantic transport events. Asian anthropogenic emissions cause less than 2 ppbv enhancement of surface ozone over Europe. The North



**Figure 19.** Mean ozone concentration enhancements in June–August 1997 at 45°N due to anthropogenic emissions in (top) North America, (middle) Europe, and (bottom) Asia, as determined from sensitivity simulations with these emissions shut off. The dashed lines indicate continental boundaries.

American influence on Europe is driven by the long-range transport of ozone rather than of its precursors, and includes comparable contributions from direct transatlantic transport of ozone in the boundary layer and subsidence from the free troposphere over Europe. Our model results suggest that 20% of the violations of the European Council ozone standard (55 ppbv, 8-hour average) in most of Europe in the summer of 1997 would not have occurred in the absence of North American anthropogenic emissions. North American influence on surface ozone in Europe is maximum when surface ozone concentrations are about 50–60 ppbv (i.e., at the threshold of the standard). North American influence is less during acute high- $O_3$  episodes associated with regional stagnation.

[48] **Acknowledgments.** We thank Mathew Evans for providing air trajectories at Mace Head. We would like to acknowledge the helpful

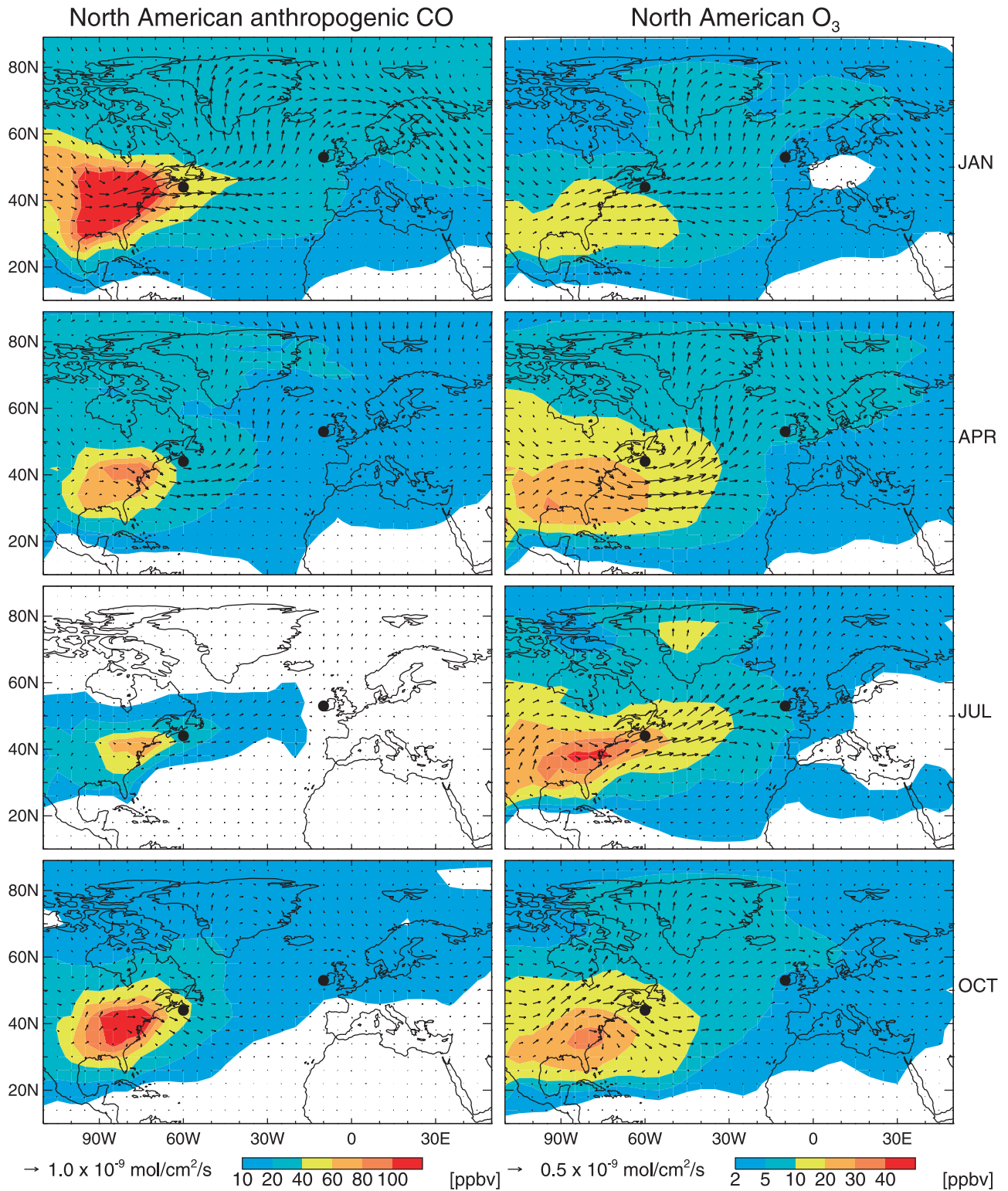
comments from two anonymous reviewers. This work was funded by the Office of Global Programs of the National Oceanic and Atmospheric Administration (NOAA) and by the National Aeronautics and Space Administration (NASA) Atmospheric Chemistry Modeling & Analysis Program. The Mace Head observations were supported by grants from the UK Department of the Environment (now DEFRA) (contracts PECD 7/10/154, and EPG 1/1/37 and 1/1/82), and by grants from the Upper Atmospheric Research Program of the National Aeronautics and Space Administration (NASA) (grants NAGW-732, NAG1-1805, and NAG5-3794 to MIT).

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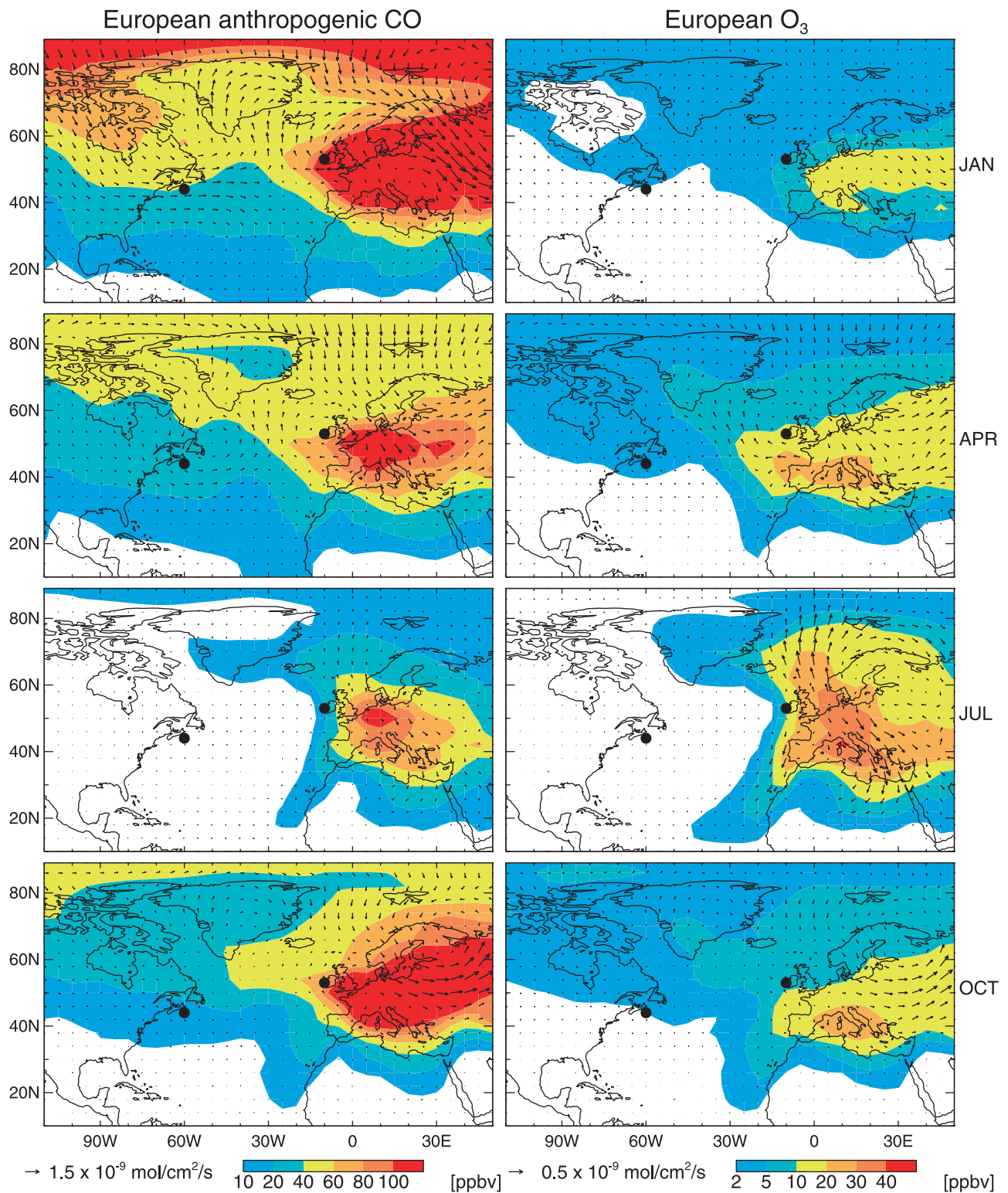
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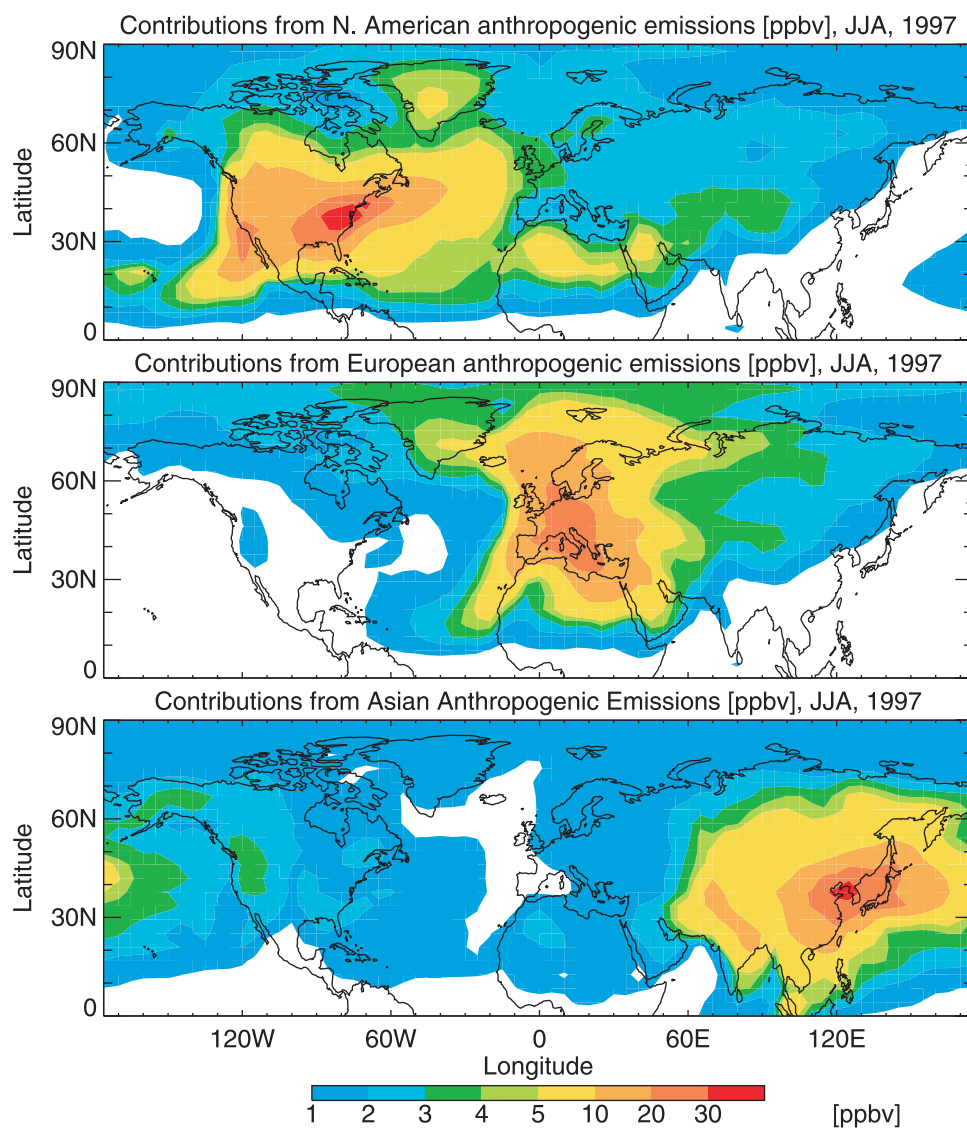
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**Figure 3.** Simulated monthly average fluxes at 0–3-km altitude of (left) North American anthropogenic CO and (right) North American ozone for January, April, July, and October 1997. Contours are concentrations in the lowest model layer (0–100 m above the surface). Solid circles indicate the locations of Mace Head and Sable Island.



**Figure 4.** Simulated monthly average fluxes at 0–3-km altitude of (left) European anthropogenic CO and (right) European ozone for January, April, July, and October 1997. Contours are concentrations in the lowest model layer (0–100 m above the surface). Solid circles indicate the locations of Mace Head and Sable Island.



**Figure 16.** Sensitivity of simulated surface ozone concentrations in June–August 1997 to anthropogenic emissions in the individual continents. The plots show the mean decreases in concentrations when anthropogenic emissions from (top) North America, (middle) Europe, and (bottom) Asia are shut off separately. “Surface ozone” refers to the lower mixed layer (0–100 m), i.e., the lowest layer of the model.