Sources of ozone over the North Atlantic and trans-Atlantic transport of pollution: a global model perspective

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Introduction

There is considerable interest in understanding the linkages between regional ozone pollution and global atmospheric chemistry because of the implications for the oxidizing power of the atmosphere, greenhouse radiative forcing by ozone, and intercontinental transport of pollution. Particular focus over the past decade has been placed on the North Atlantic Ocean (NAO) atmosphere through a series of field programs [*Parrish*, this issue]. We have recently applied the GEOS-CHEM global 3-D model of tropospheric chemistry [*Bey et al.*, 2001a] driven by assimilated meteorological observations for 1993-1997 from the NASA Data Assimilation Office (DAO) [*Schubert et al.*, 1993] to a detailed analysis of observations from the 1997 North Atlantic Regional Experiment (NARE) [*Fehsenfeld et al.*, 1996], the Atmospheric Chemistry Studies in the Oceanic Environment (ACSOE) [*Sturges et al.*, 1996], and the 2nd Atmospheric Characterization Experiment (ACE-2) [*Raes et al.*, 2000], as well as ozonesonde and surface measurements over the NAO. Model budget analyses, tagging of species by source region, and sensitivity simulations are offering new insights on the sources of tropospheric ozone over the NAO and the trans-Atlantic transport of pollution. This work is being prepared for publication in the Journal of Geophysical Research [*Li et al.*, 2001a, b]. We present here a few of the major results.

Sources of ozone over the North Atlantic

There has been long-standing debate over the contributions of anthropogenic pollution and transport from the stratosphere to the tropospheric ozone budget over the NAO. *Oltmans et al.* [1992, 1994] have argued for a major stratospheric influence on surface ozone at Bermuda in spring on the basis of strong subsidence indicated by trajectory analyses. However, the relationship of subsiding trajectories to the actual origin of ozone is ambiguous [*Moody et al.*, 1995]. Other studies have found evidence for a major contribution from North American pollution to the springtime ozone maximum at Bermuda [*Dickerson et al.*, 1995; *Prados et al.*, 1999]. A positive O_3:CO correlation at Sable Island and other Canadian Atlantic sites in summer was reported by *Parrish et al.* [1993] and implies a dominance of pollution over stratospheric influence at least for that season. Negative O_3:CO correlations in winter at the same sites have been attributed to ozone titration by NO_x and hydrocarbons [*Parrish et al.*, 1998]. Similar correlations between ozone and CO have been observed at Mace Head on the western coast of Ireland [*Derwent et al.*, 1994].

The observed O_3:CO correlations at Sable Island and Mace Head are well simulated by our model (Figure 1), providing support for the model representation of photochemistry and transport over the NAO. Further comparison of observed and simulated time series of ozone and CO concentra-

tions at Bermuda and Mace Head in 1996-1997 are shown in Figure 2; the success of the model in capturing both background values and high events is apparent. In that Figure we have decomposed model ozone and CO into contributions from different source regions by using tagged tracers [*Wang et al.,* 1998; *Bey et al.,* 2001b]. We find that most of the surface ozone observed at Bermuda in spring originates from the lower troposphere over North America, while transport from the upper troposphere/lower stratosphere plays only a minor role (Figure 2a). Similarly, our simulations indicate that most of the ozone observed at Mace Head in spring is produced in the lower troposphere over Europe (Figure 2b), and this is further supported by decomposition of the time series of CO concentrations in the model (Figure 2c). The "other" contribtions in Figure 2a and 2b are mainly from ozone productions in the lower troposphere over the NAO. Considering that the ozone precursors over the NAO are largely from North America and Europe, the anthropogenic influence from North America on surface ozone in Bermuda and from European on surface ozone in Mace Head may be even larger.

Prospero et al. [1995] found that summertime ozone at Izana in the Canary Islands (28°N, 16°W, 2.4 km) is positively correlated with ^{7}Be aerosols, and negatively correlated with ^{210}Pb aerosols, and viewed these relationships as evidence for a high-altitude source of ozone, possibly from the stratosphere. We examined the ability of the GEOS-CHEM model to reproduce these correlations, using the radionuclide simulation previously reported by *Liu et al.* [2001]. We find that simulated ozone at Izana is correlated with ^{7}Be (r=0.80; slope=0.39) and anti-correlated with ^{210}Pb (r=0.50, slope=-0.01) [*Li et al.,* 2001b]. Tagged ozone tracer simulations show that these correlations are mostly driven by ozone production in the middle and upper troposphere, with transport from the stratosphere contributing less than 5 ppbv ozone at the site.

There has been considerable recent interest in the effect of ship NO_x emissions on ozone production over the NAO [*Lawrence and Crutzen,* 1999; *Kasibhatla et al.,* 1999]. In our standard simulations we use ship emission estimates from the Global Emissions Inventory Activity (GEIA) [*Benkovitz et al.,* 1996], which are sufficently low that ships play little role in the ozone budget. A recent ship emission inventory by *Corbett et al.* [1999] indicates much larger values, implying a large influence from ships on ozone and OH over the NAO [*Lawrence and Crutzen,* 1999]. *Kasibhatla et al.* [1999] compared the 1997 NARE aircraft observation statistics for NO and NO_y with results from a global 3-D model driven by GCM meteorology, and concluded that the high ship emissions of *Corbett et al.* [1999] were inconsistent with the aircraft observations. By comparing to specific 1997 NARE observations removed from continental influence, our simulations show indeed that when the ship emission inventory of *Corbett et al.* [1999] is included, the marine boundary layer NO and NO_y concentrations are overestimated by a factor of 2; ozone concentrations are increased by about 10 ppbv. We conclude that the aircraft observations were inconsistent with the representation of the high ship NO_x emissions of *Corbett et al.* [1999] in their model.

Trans-Atlantic transport of pollution

Analyses of observations have shown evidence for long-range transport of North American ozone pollution to the middle and upper troposphere over Europe, but little effect on the surface [*Stohl and Trickl,* this issue]. Tagging of ozone and CO enable us to diagnose trans-Atlantic transport of pollution in the GEOS-CHEM simulation. We find that ozone produced in the lower troposphere (>700 hPa) over North America contributes on average about 5 ppbv to the surface ozone at Mace

Head, but as much as 10-15 ppbv during some transatlantic transport events. Figures 2b and 2c show such events on March 23-26 and April 26-29, 1997. We find that these trans-Atlantic pollution transport events are less frequent in summer than in other seasons [*Li et al.*, 2001b].

A remarkable result of our simulation is the occasional occurrence of trans-Atlantic transport of European pollution to North America. We show in Figure 3 one such event during June 3-6, 1997. On June 2-3, a high pressure system formed between the Hudson Bay and the North Sea along 60°N while a low pressure system was developing between Newfoundland and the eastern NAO along 45°N. Between the two systems a low level (below 2 km) easterly flow was established, bringing European pollution directly to the northeastern North America. This easterly flow stopped on June 4 as the low pressure system moved eastward. However, the European pollution cloud persisted over the northeastern North America until June 8, with large effects on simulated ozone and CO at Sable Island (Figure 4). The "other" contribution in Figure 4a is mainly from ozone production in the lower troposphere over the NAO, which was particularly large during the June 3-6 event. Under the flow pattern discussed above, it is likely that the "other" contribution during the June 3-6 event were due to European pollution being transported to the NAO and subsequent ozone production. As a result, the European influence during the June 3-6 event may be even larger. Unfortunately, no observations were available for that period, and the anomalous circulation pattern did not recur for the remainder of the summer. However, we find similar events occurred almost every spring and sometimes fall for the time period that we simulated, i.e., 1993-1997.

In sensitivity simulations with either North American or European fossil fuel emissions shut off, we find that anthropogenic sources in North America enhance July mean ozone concentrations in surface air over Europe by 2-4 ppbv, with maxima over the British Isles (Figure 5, left panel). Greater North American influence is actually found over North Africa and the Middle East because of the deep mixing in these arid regions [*Li et al.*, 2001c]. Anthropogenic sources in Europe enhance July surface ozone over North America by only 1-2 ppbv (Figure 5, right panel).

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Figures

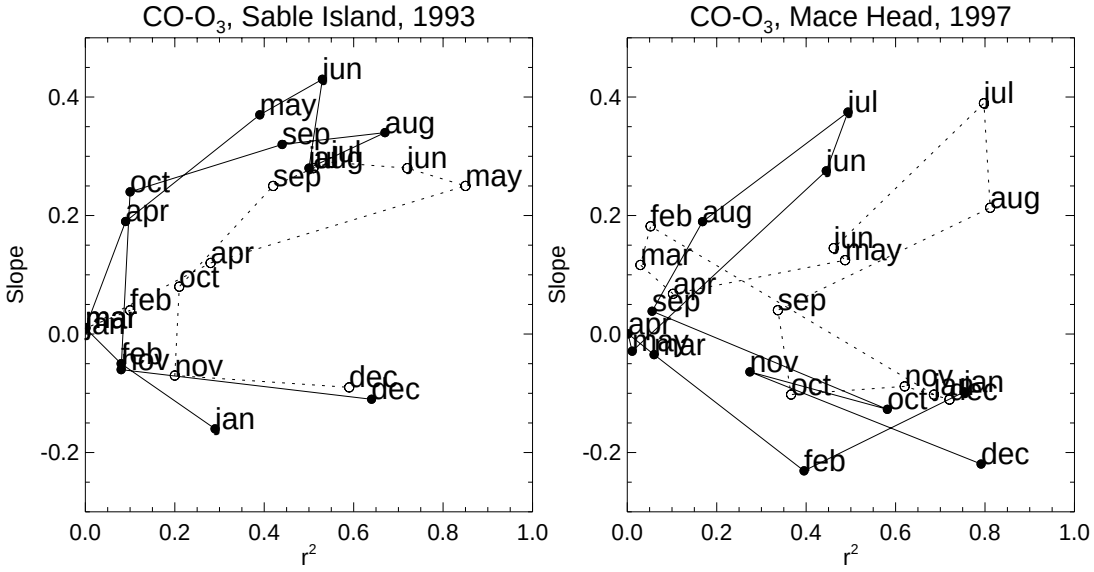

Figure 1. Correlations between ozone and CO concentrations at Sable Island on the east coast of Canada (44°N, 60°W) and at Mace Head on the west coast of Ireland (53°N, 10°W). The figure shows monthly correlation coefficients and slopes in the observations (solid circles) and in the GEOS-CHEM model (open circles). The Sable Island observations are for 1993 [*Parrish et al.*, 1998] and the Mace Head observations are for 1997 (Peter Simmonds and Gerard Spain, personal communication). Model results are for the corresponding years.

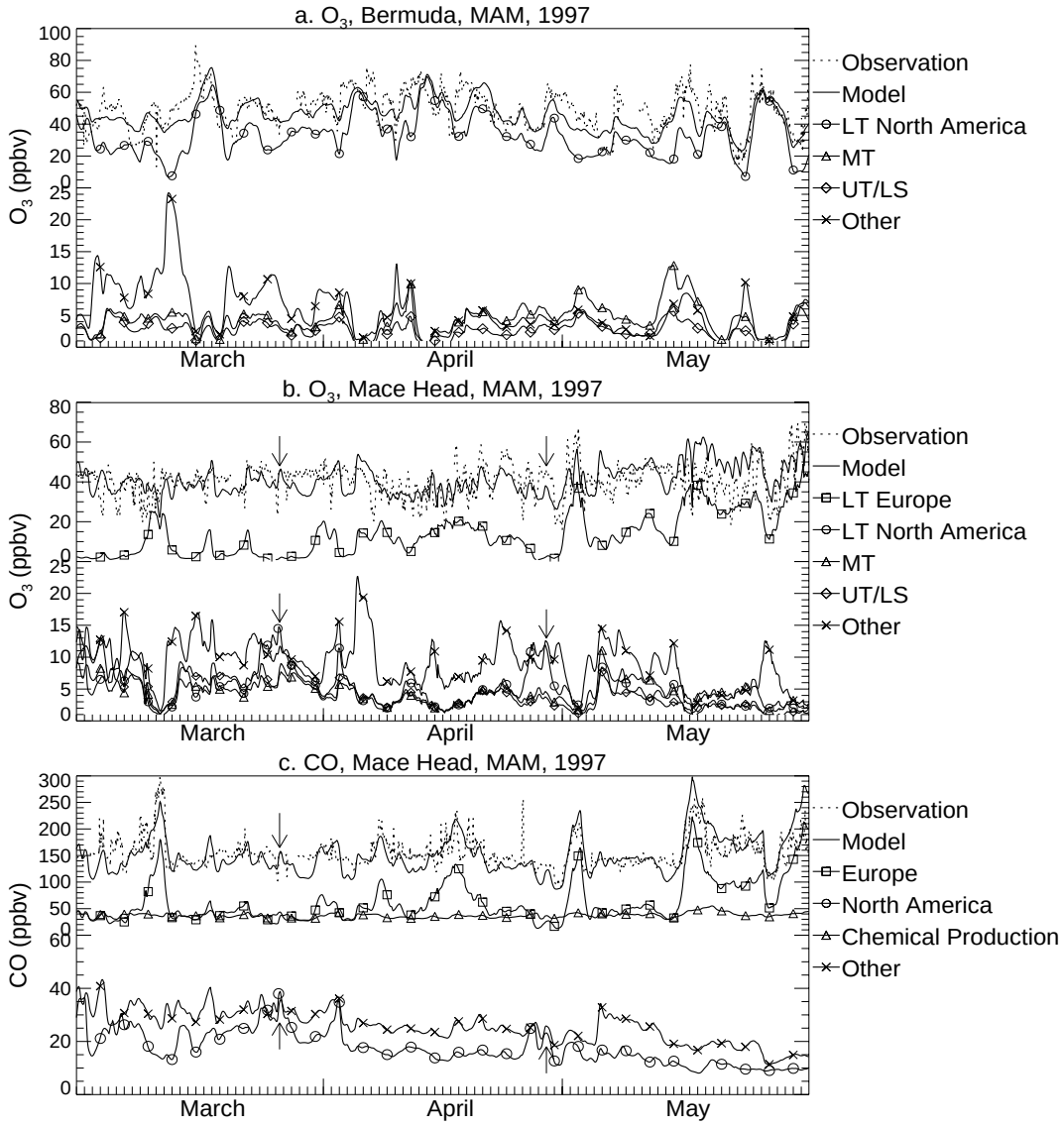

Figure 2. Observed (dotted line) and simulated (solid line) time series of surface concentrations of (a) ozone in spring 1996 at Bermuda and (b) ozone in spring 1997 at Mace Head, and (c) CO in 1997 at Mace Head. Contributions from different sources are isolated in the model with tagged ozone and CO tracers [*Wang et al.*, 1998; *Bey et al.*, 2001b]. In this manner, total ozone concentrations are decomposed into contributions from production in (1) the upper troposphere (< 400 hPa) and lower stratosphere (UT/LS); (2) the middle troposphere (MT; 400-700 hPa); and (3) the lower troposphere (LT; > 700 hPa) over North America and Europe separately. Total CO concentrations are decomposed into contributions from direct anthropogenic emissions in different continents and chemical production within the atmosphere. Ozone data for Bermuda were provided by Samuel Oltmans. Ozone and CO data for Mace Head are provided by Peter Simmonds and Gerard Spain, respectively. The arrows highlight North American pollution events at Mace Head discussed in the text.

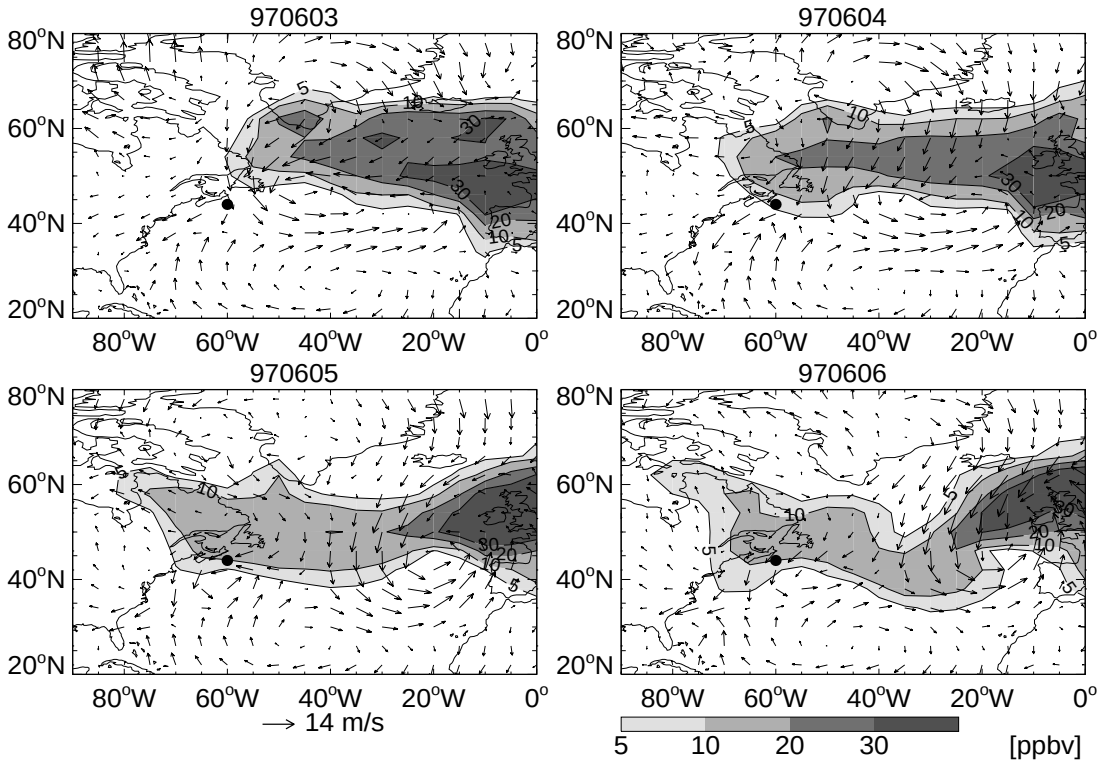

Figure 3. Simulated surface concentrations on June 3-6, 1997 of the European pollution ozone tracer produced in the lower troposphere over Europe on June 3-6, 1997. GEOS surface wind fields are also shown. The location of Sable Island is indicated by the solid circle.

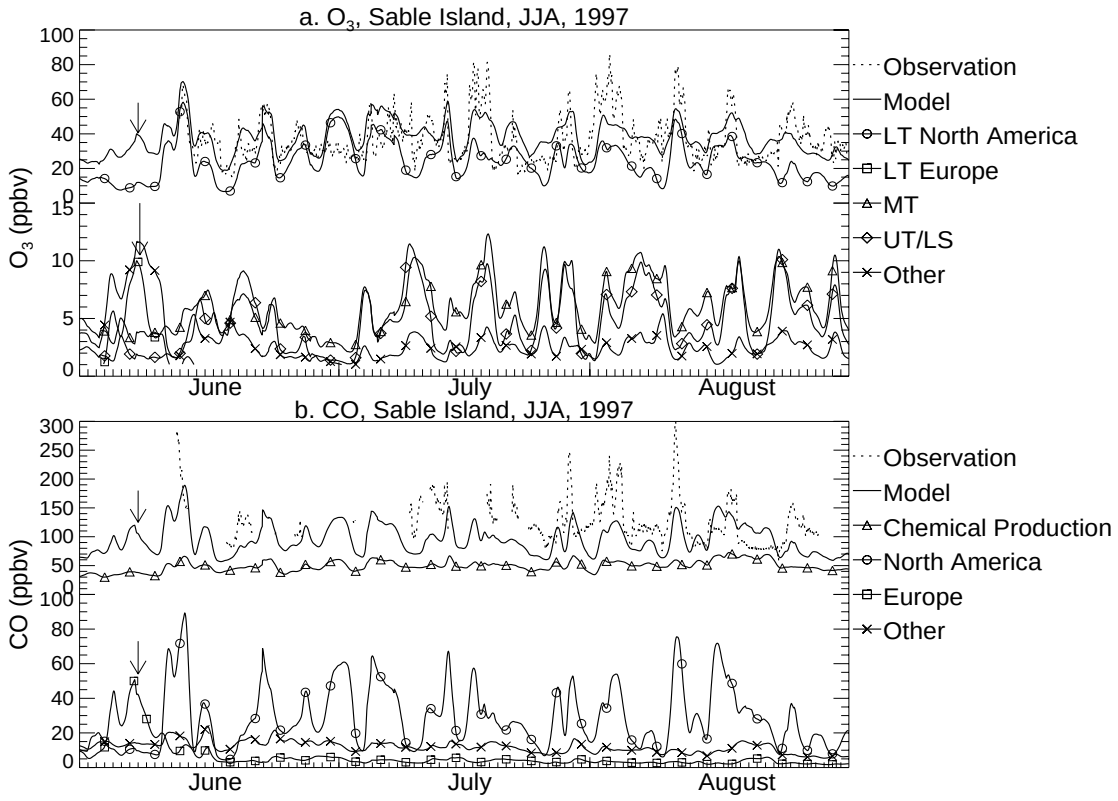

Figure 4. Observed (dotted line) and simulated (solid line) time series of surface concentrations of ozone and CO at Sable Island in the summer of 1997. Contributions from different sources are isolated in the model with tagged tracers as in Figure 2. The arrows indicate the European pollution event in the model on June 7-8, 1997. Observations are from David Parrish (personal communication).

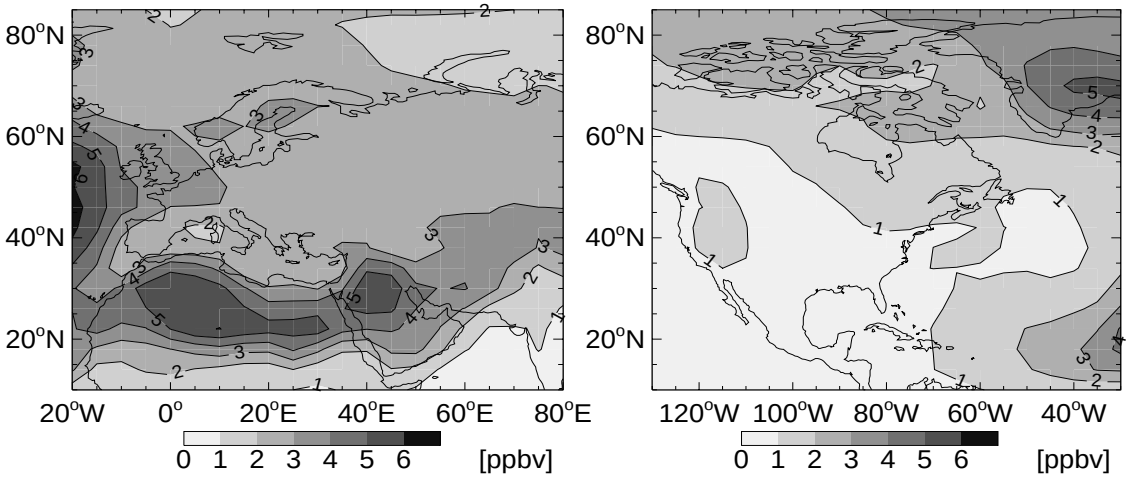

Figure 5. Decrease in simulated monthly mean ozone concentrations in surface air in July over Europe when North American fossil fuel emissions are shut off (left panel) and over North America when European fossil fuel emissions are shut off (right panel).