The Contribution of Global Transport on North American Carbon Monoxide Concentrations

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B.S. Geography 2007

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-18 May 2007-

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List of Acronyms

CMDL	Climate Monitoring and Diagnostic Laboratory
EDGAR	Emission Database for Global Atmospheric Research
EPA	Environmental Protection Agency
GEIA	Global Emission Inventory Activity
GFDL	Geophysical Fluid Dynamics Laboratory
ICARTT	International Consortium for Atmospheric Research on Transport and Transformation
INTEX-NA	Intercontinental Chemical Transport Experiment - North America Phase A
ITCZ	Intertropical Convergence Zone
LRTAP	Long-Range Transport of Atmospheric Pollutants
MODIS	Moderate Resolution Imaging Spectroradiometer
MOZART	Model for Ozone and Related Tracers
NASA	National Aeronautics and Space Administration
NCAR	National Center for Atmospheric Research
NCEP	National Center for Environmental Prediction
NOAA	National Oceanic and Atmospheric Administration
TFHAP	Task Force on Hemispheric Transport of Air Pollution

Acknowledgments

I must thank Tracey Holloway, my mentor at the Center for Sustainability and the Global Environment (SAGE), for introducing me to this field of research and advising me through my education and professional developments. Thank you to Jack Williams at the UW-Madison Department of Geography for his support as my advisor. I also appreciate Arlene Fiore and Larry Horowitz for mentoring me during an internship at the Geophysical Fluid Dynamics Laboratory. I would like to thank Yuanyuan Fang and Claus Moberg for their support, aide, and research assistance. Funding for this project was provided by the National Oceanic and Atmospheric Administration's Ernest F. Hollings Undergraduate Fellowship from 2005-2007. Lastly, thank you to my family and friends for their unending support and encouragement.

I. Abstract

Carbon monoxide, CO, is produced by natural and anthropogenic processes including biomass burning and fossil fuel usage and affects atmospheric chemistry through its roles as a sink for the hydroxyl radical (OH) and as a precursor to ozone. As the primary atmospheric sink for OH, which is responsible for chemically destroying numerous air pollutants, CO concentrations impact the concentrations of other such pollutants. Here we use CO as a tracer for polluted air masses by examining the transport of CO both to and from North America.

CO is an ideal tracer for atmospheric and climate modeling because it is well understood and well captured due to its simple chemistry and long lifespan. By employing MOZART, a numerical global tropospheric chemistry model, we seek to address the nature of air pollutant transport and establish the role of transport on regional North American CO concentrations. We find that the greatest intercontinental transported CO occurs on days when the overall carbon monoxide concentrations are low to moderate. In addition, carbon monoxide concentrations increase eastward, reflecting the different regional impacts of emissions.

We also define three main transport pathways of CO over North America and identify specific episodic flux events by comparing model results to INTEX-NA flight observations taken the summer of 2004 in cooperation with NASA, NOAA, and the ICARTT campaign. The main pathways of CO transport over North America are eastward, aloft import from Asia; northward, surface import from Africa, attributed to heavy biomass burning in the summer; and eastward export from North America, at the surface and aloft.

Understanding these pathways for CO transport and the regional impacts is a key step towards understanding how polluted air masses evolve and can provide insight into the extent to which local air quality is influenced by intercontinental transport.

II. Introduction

Carbon monoxide, CO, is an atmospheric pollutant that is both well understood and regulated. Because the chemistry of carbon monoxide is relatively simple, it is an easy species to capture with an atmospheric chemistry numerical model. The main reaction of most import is between carbon monoxide and hydroxyl, OH, whereby OH acts as a sink for CO.

$$CO + OH \rightarrow CO_2 + H$$

This is significant as OH is a highly reactive sink for numerous other atmospheric constituents, like sulfur dioxide and nitrogen oxides. Therefore the presence of CO plays a critical role in controlling OH concentrations in the atmosphere and is a precursor to ozone [*Jacob,* 1999]. Like ozone, high levels of carbon monoxide can lead to health effects, but on the global scale, levels of CO are much smaller.

Carbon monoxide is produced by biogenic and anthropogenic processes and has a lifetime of about two months in the atmosphere [*Jacob,* 1999]. Biomass burning accounts for approximately 25% of carbon monoxide emissions. Fossil fuel use, a major anthropogenic source, makes up 20% of the CO emissions. Oxidation of methane and other hydrocarbons accounts for about 44% of CO emissions and is the largest biogenic source [*Jacob*, 1999]. Figure 1 illustrates these sources and sinks of CO.

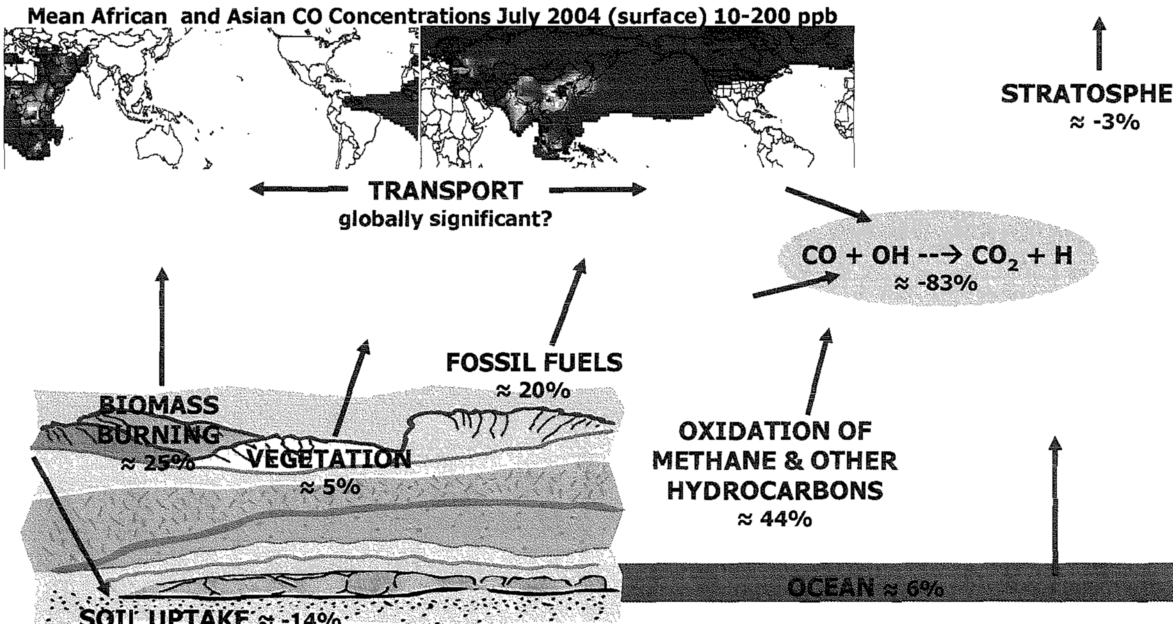


Figure 1: Carbon Monoxide Schematic of Sources and Sinks (adapted from Jacob 1999)

The global distribution of carbon monoxide has been modeled in previous studies using global models such as *Pinto et al,* [1983]; *Saylor and Peters,* [1991]; *Muller and Brasseur*, [1995]; *Allen et al,* [1996]; *Berntsen and Isakson,* [1997]; *Wang et al,* [1998]; *Kanakidou et al,* [1999]; *Holloway et al,* [2000]; *Bey at al,* [2001] and [2002]; *Kiley* et al, [2003]; and *Shindell et al*, [2006]. Each study has used carbon monoxide in various ways from understanding the global distribution of carbon monoxide [*Holloway et al,* 2000] to studying it as part of Asian chemistry [*Bey et al*, 2001]. Because CO has been studied extensively, its behavior is well known. In addition, its long lifetime enables global models to use it is an ideal tracer and indicator for polluted air masses. In this way, CO can be used as an aid to understanding the evolution of air masses and the global transport of air pollution [*Holloway et al,* 2000].

Carbon Monoxide. Carbon monoxide has been evaluated at various temporal and spatial resolutions over North America, Europe, and Asia. Studies have shown that carbon monoxide concentrations have seasonal variability due in part because carbon monoxide builds up over winter, causing an annual maximum in concentrations [*Wild et al*, 2004; *Bey et al*, 2001 and 2002; and *Yienger et al*, 2000]. European CO concentrations have a seasonal peak in the spring and minimum in the summer also [*Wild et al*, 2004]. In modeling, carbon monoxide emissions are assumed to be constant for the entire year, which is necessary given the lack of emission inventories. Therefore, the daily variability in modeled carbon monoxide concentrations over North America is largely driven by daily variations in meteorology and general circulation [*Fiore et al,* 2003]. In addition, previous studies have found higher surface concentrations of carbon monoxide at higher latitudes due to rapid oxidation with high concentrations of OH in the low latitudes [*Bey et al,* 2001]. *Bey et al,* [2001] also found greater CO concentrations at higher latitudes over northwestern China. It is expected that the rapid industrialization of eastern Asia will have important impacts and changes on not only carbon monoxide concentrations but also on atmospheric chemistry in the future [*Bey et al*, 2001].

Intercontinental Transport. Given the changing global distribution of carbon monoxide (figure 2), intercontinental transport becomes important to understand as international communities are working to

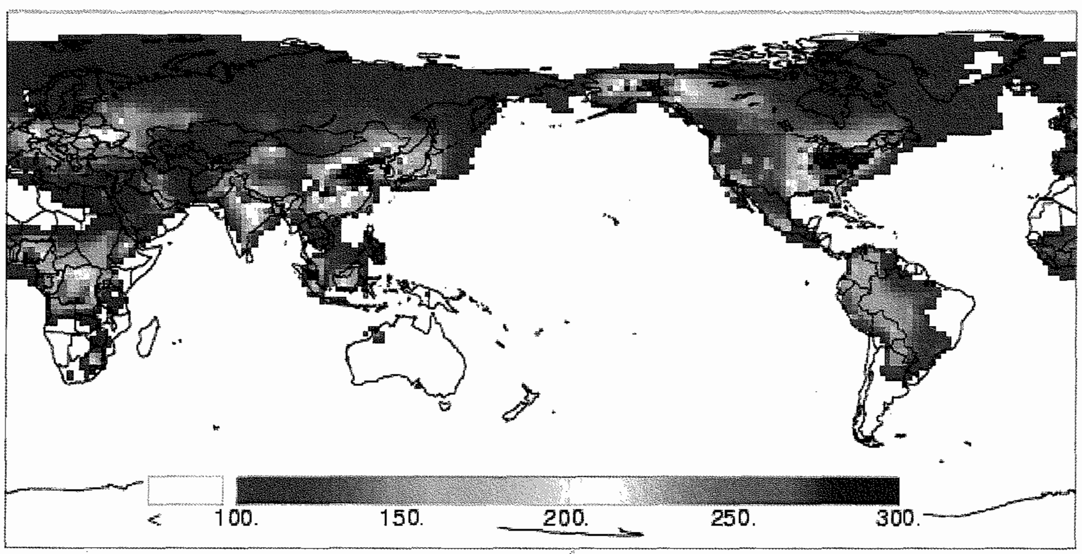


Figure 2: Mean Surface Carbon Monoxide Concentrations, July 2004 (ppb)

increase knowledge and create policy frameworks. Unfortunately, while carbon monoxide may be well understood, intercontinental transport of air pollution is not. However, the scientific and policy communities are showing increasing interest [*Fiore et al*, 2003]. New scientific organizations have formed recently to study and understand the transport of atmospheric species. For example, both the Task Force on Hemispheric Transport of Air Pollution (TFHAP) and the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) are recent developments, designed to study transport of air pollution.

Among the species that have been studied for its global transport are ozone (O_3), mercury (Hg), carbon monoxide (CO), and particulate matter (PM) and aerosols as they relate to dust storms [*Bey et al, 2002*]. General transport mechanisms have been identified, which include warm conveyer belts that transport pollution aloft from Asia to North American and from North America to Europe. Advection is a mechanism in the boundary layer that causes pollutants to transport from Europe to Asia and occasionally from North America to Europe [*Fiore et al*, 2003].

Like the seasonal variability of carbon monoxide levels, intercontinental transport from Asia reaches a maximum during the spring with the creation of strong cold fronts due to the Tibetan Plateau, which causes uplift of Asian pollutants [*Fiore et al*, 2003 and *Bey et al,* 2001]. Asian carbon monoxide can take from 5-10 days to cross the Pacific Ocean and reach North America [*Wilkening et al,* 2000]. Asian CO concentrations are transported as far as North America, Siberia, and Eastern Europe as observed by fossil fuel and biomass burning signatures [*Bey et al*, 2001].

With increasing interest of the global community on atmospheric species transport, the need for more understanding is apparent. By utilizing a global model and observations, we seek to understand North American carbon monoxide concentrations and establish the role of transported CO on North America. It is has been shown that Asian carbon monoxide exceeds North American carbon monoxide in the free troposphere over North America [*Yienger et al*, 2000]. Similarly, *Holloway et al*, [2000] found that at the 500 mbar level (the free troposphere), higher wind speeds and longer CO lifetimes permits transport over greater distances. North American carbon monoxide concentrations shift northeastward, suggesting that North American CO may influence Europe and Greenland [*Li et al*, 2005].

This study hopes to add to the understanding of intercontinental transport of CO by (one) studying modeled carbon monoxide concentrations at the regional scale over North America and (two) identifying specific transport mechanisms of carbon monoxide into and out of the eastern United States. In doing so, we hope to provide a qualitative overview of carbon monoxide transport and its impacts at the regional scale over North America.

III. Methodology

Three methods are used in this study to fully understand global transport of CO and its regional impacts on North America. By combining a global model with regional analysis and observations from the eastern United States, we study many features of global CO transport.

Modeling. Carbon monoxide works ideally for such a study as it is readily modeled and is monitored by the global community [*Holloway et al,* 2000]. A variety of models have been used to interpret observations and identify intercontinental transport pathways, including three-dimensional (3-D) global chemical transport models, which is the type we utilize in this study [*Fiore et al,* 2003]. In this way, we model carbon monoxide transport over North America first by quantifying regional contributions. We compare tagged CO more specifically with daily observations from over the eastern United States as well.

The Model for Ozone and Related Tracers (MOZART) is used at the regional scale to understand the development and patterns of global transport of CO over North America. MOZART, a global, numerical tropospheric model (figure 3), was created by scientists at the National Center for Atmospheric Research (NCAR), the Max Planck Institute for Meteorology, and the Geophysical Fluid Dynamics Laboratory (GFDL) [*Horowitz et al*, 2003].

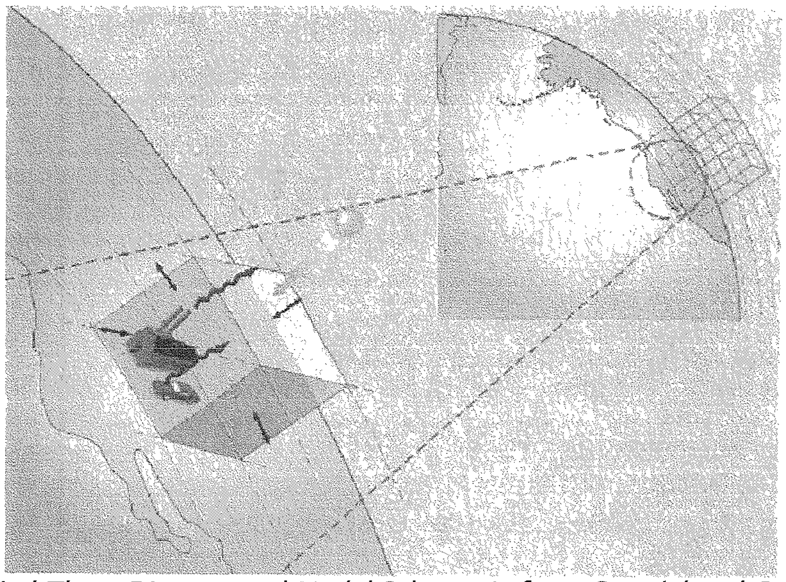


Figure 3: Global Three Dimensional Model Schematic from Graedel and Crutzen, 1997

MOZART-2 uses National Center for Environmental Prediction (NCEP) meteorology and emissions based on inventories from the Emissions Database for Global Atmospheric Research (EDGAR) v2.0, the Global Emission Inventory Activity (GEIA), and Hao and Liu, 1994 and Müller, 1992. With a grid resolution of about 1.5°, MOZART is appropriately suited for applications in regional studies.

MOZART has been used in the past for studies with CO and *Horowitz et al* [2003] found that global budgets of tropospheric ozone CO are consistent with recent studies. *Horowitz et al* [2003] also found in their evaluation of MOZART, version 2, that mean simulated CO concentrations agree well with observations at most sites. MOZART, while capturing carbon monoxide well, has shown to misrepresent the biomass burning seasonality of the tropics, which accounts for about 50% of direct CO emissions. MOZART misrepresents the seasonality of CO in northern latitudes as well [*Holloway et al*, 2000]. MOZART-2 also accurately simulates global CO budgets based on other model studies of CO budgets by *Hauglustaine et al*, [1998]; *Granier et al*, [2000]; *Holloway et al*, [2000], and *Bey et al*, [2001].

Regional analysis. To analyze the nature of globally transported CO on North America, we examined nine regions in the United States, selected to capture the major population centers with approximately the same size (table 1 and figure 4).

Region	Lat (°N)	Lon (°W)
Northwest	42 - 49	120 - 125
Northern CA	37 - 42	120 - 125
Southern CA	32 - 37	120 - 125
Midwest	40 - 47	80 - 90
Mountain West	35 - 40	105 - 115
Gulf Coast	30 - 35	85 - 95
Northeast	40 - 45	70 - 80
Mid-Atlantic	35 - 40	75 - 80
Southeast	30 - 37	80 - 85

Table 1: Latitudes and Longitudes of nine North American Regions

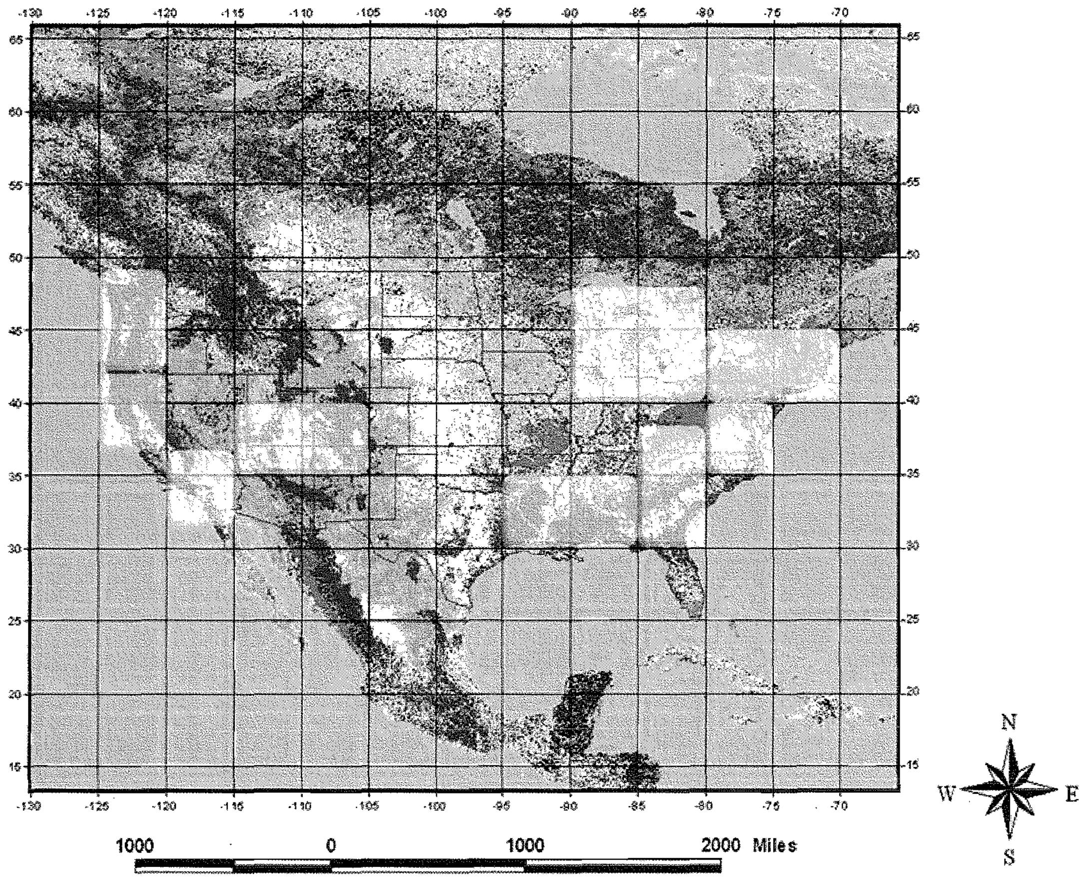


Figure 4: Land cover map of North America with nine regions highlighted, from Moberg, 2007

For each region, carbon monoxide concentrations from Asia, Europe, and North America were plotted daily for 2001 and 2002. To do so, we run MOZART simulations with a specific region "turned off" or not included. These runs are subtracted from MOZART simulations of total CO. Three regions, Asia, Europe, and North America, were calculated this way. MOZART simulated carbon monoxide concentrations were averaged over each defined U.S. region. In addition, total Asian and European CO, not averaged over the area, was also plotted against total daily CO for each of the nine U.S. regions. This enables visualization of global transport at the regional level.

Model and observation comparison in the eastern United States. The last method employed in this study was to compare MOZART simulated tagged CO with observations over the eastern United States in order to enhance our regional understanding of global transport but also to evaluate the model. We ran MOZART at the daily and hourly level for the summer of 2004 with the purpose of being able to compare model results with the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) data. ICARTT was created in 2003 to study regional air quality, intercontinental transport, and radiation balance [*Fehsenfeld et al,* 2006]. ICARTT incorporates satellite, ground, and flight measurements to create a database of observations to be used in coordination with model users. Ground observations were taken in the northeastern United States, Nova Scotia, the Azores, and western Europe.

As part of ICARTT, the Intercontinental Chemical Transport Experiment - North America (INTEX-NA), a NASA supported study, conducted extensive ground and flight observations over the eastern United States. A particular focus of INTEX-NA was to quantify and characterize the inflow and outflow of pollution over North America [*Singh et al*, 2006]. The main constituents of interest are ozone, aerosols, and their precursors, and long-lived greenhouse gases (GHGs). From July 2004 to mid-August 2004, two planes, NOAA P3-B and NASA DC-8 took measurements of numerous species, among them was carbon monoxide. The goals of INTEX-NA were to understand the chemical evolution of North American outflow over the Atlantic and to quantify the transpacific transport of Asian pollution to the United States based on the understanding of the Long-Range Transport of Atmospheric Pollutants (LRTAP) [*Singh et al,* 2006]. INTEX-NA is part of a number of coordinated national and international studies and field campaigns.

INTEX-NA observations were plotted against modeled data in order to assess the nature of intercontinental transport of carbon monoxide over North America. The extensive coverage of plane observations over the eastern United States provided a unique opportunity to compare model results with numerous observations. By comparing MOZART to observations, we were able to more accurately analyze and understand the modeled results as well as evaluate the model's ability to capture CO.

IV. Sources of North American Carbon Monoxide

Carbon monoxide concentrations on the global scale tend to decrease from north to south and peak in the late winter and early spring, with a decrease in the summer [*Novelli et al*, 1994; *Wild et al*, 2004; *Bey et al*, 2001 and 2002; and *Yienger et al*, 2000]. However, due to variability in its sources and sinks, CO has significant inter-annual variation in addition to its seasonal cycle. A strong seasonal cycle occurs especially in the northern hemisphere according to studies by *Novelli et al* [1994]. Winter CO is dominated by fossil fuel emissions, which tend to pool over the eastern United States and mid-Atlantic [*Holloway et al*, 2000]. In the summer, OH concentrations are greatest, resulting in the shortest CO lifetime and lowest total concentrations.

Carbon monoxide concentrations are further influenced by emissions, which vary from year to year and season to season. Summer is the burning season in Africa, which contributes to increases in CO in the tropics. Furthermore, oxidation of methane, biogenic emissions, and biomass burning (forest fires in the western United States and Canada) cause an increase in sources in the summertime. As carbon monoxide emissions are assumed to be constant for the entire year [*Holloway et al,* 1999], the daily variability in carbon monoxide concentrations is largely driven by daily variations in meteorology and general circulation as found in *Fiore et al* [2003]. Given our understanding of global and North American carbon monoxide, we expect regional CO levels to follow a similar seasonal example and that global transport will be limited in the summer.

To examine the impacts of globally transported carbon monoxide on North America, given this context, we plot daily carbon monoxide concentrations from 2001 and 2002 (figures 5 a-i and 6 a-i) with carbon monoxide from North America (green), Asia (blue), Europe (red), and other (peach). Other sources of carbon monoxide include unspecified regional sources and the formation from oxidation with methane, isoprene, and with non-methane hydrocarbons. Each of nine regions, the Northwest, Northern California, Southern California, Midwest, Mountain West, Gulf Coast, Northeast, Mid-Atlantic, and Southeast are plotted next to each other for ease of analysis.

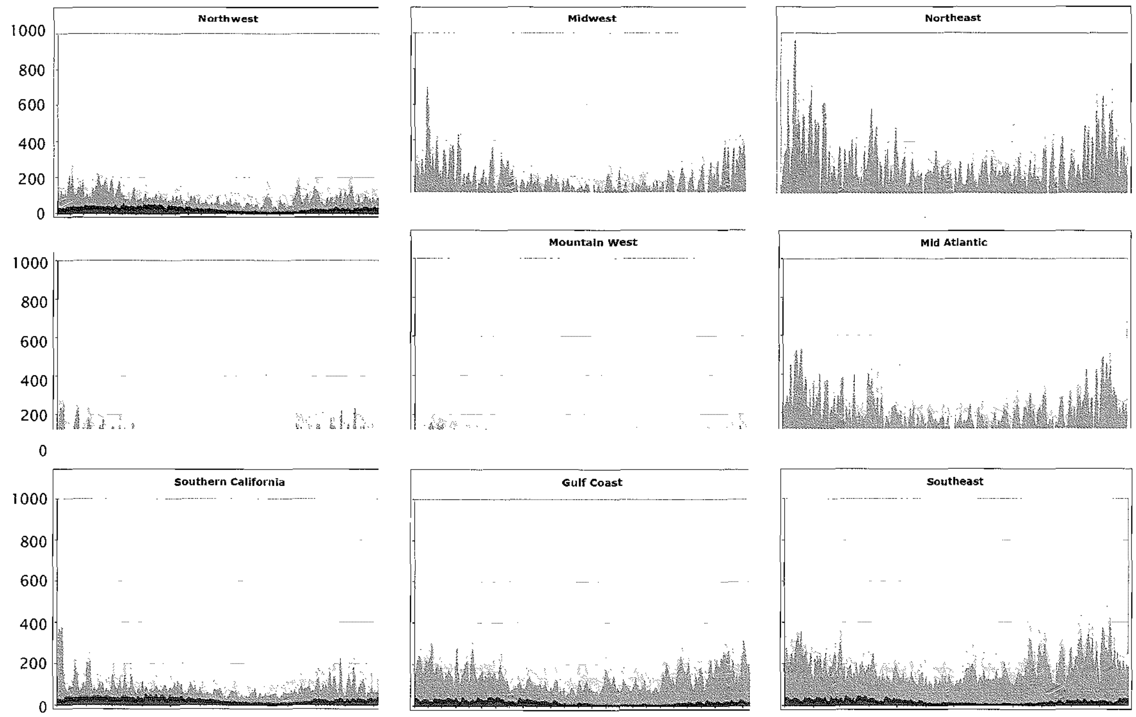


Figure 5a-i: Daily Mean Simulated CO over North America in 2001

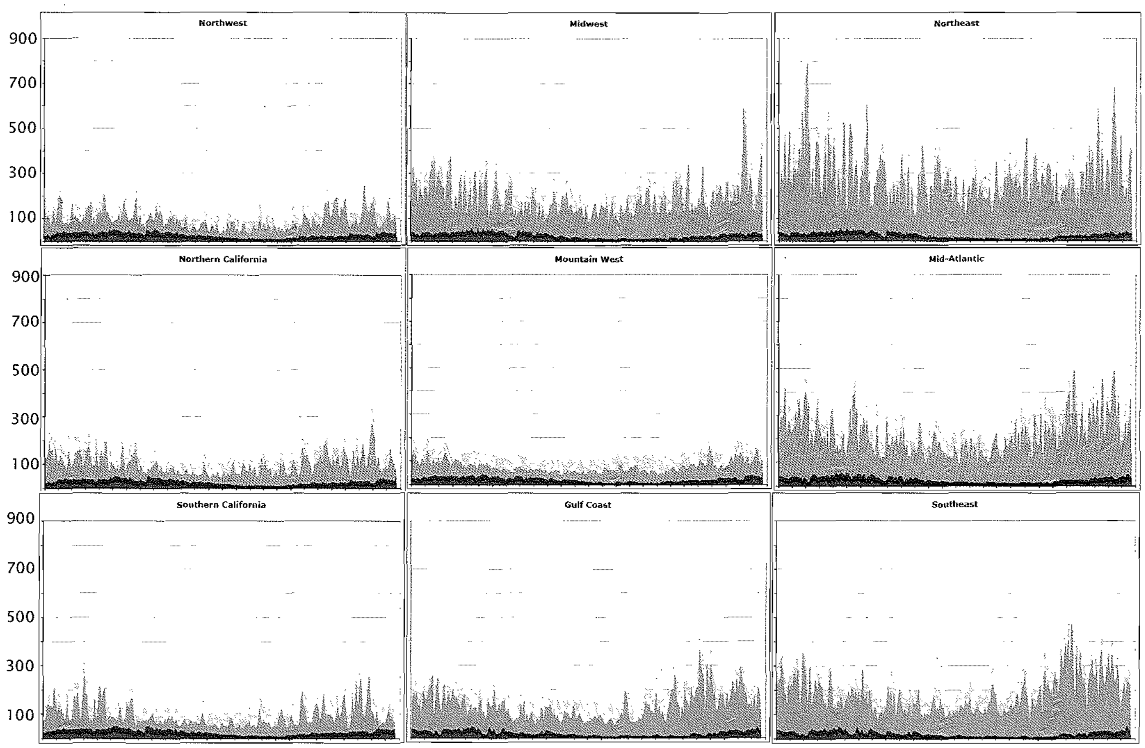


Figure 6a-i: Daily Mean Simulated CO over North America in 2002

In general, each region witnesses a seasonal trend in CO transport from Asia and Europe, with a minimum in the summer. In addition, there is an increase of total carbon monoxide concentrations eastward over North America. Due to minimal inter-annual variability in CO between 2001 and 2002, we average the two years together for analysis. The Atlantic coast sees higher CO concentrations than the Pacific coast, and Asian and European contributions vary little between regions, indicating the strong influence of local CO emissions and sources. To get a more detailed understanding of the impact of intercontinental transport of CO on North America, we analyze each region more closely.

Northwest. The mean daily carbon monoxide concentration for the Northwest is 148.4 ppb. The mean North American, Asian, and European CO concentration over the Northwest is 65.9 ppb, 16.4 ppb, and 13.3 ppb respectively. Both years see a strong seasonal pattern in the total and North American CO concentrations as well for the Northwest (figures 5a and 6a).

Northern California. The mean daily carbon monoxide concentration for Northern California in is 155.8 ppb, a little greater than in the Northwest. The mean daily North American, Asian, and European CO contribution is 74.4 ppb, 16.1 ppb, and 12.6 ppb respectively. Asian and European contribution to Northern California CO concentrations is greater in the late winter and spring months than in the fall and early winter months. *Yienger et al,* 2000 also identifies peak Asian CO concentrations in early spring with Asian transport slowing by June. In April of both years, a maximum in foreign transport occurs simultaneously with a minimum of North American and total CO concentrations (figures 5b and 6b), indicating greater transport contributions and events occurring when the overall total CO is low.

Southern California. The total daily mean concentration of carbon monoxide over Southern California is 146.4 ppb. The total CO is less than the other two Pacific Coast regions despite containing the large urban center, Los Angeles. The greater concentrations north of Los Angeles, results from small OH concentrations further north, permitting larger concentrations and longer CO lifetimes. In LA, which is farther south, OH concentrations are greater, which rapidly breaking down CO in the atmosphere, resulting in a lower CO concentration despite the urban fossil fuel CO emissions. North American, Asian, and European daily mean contribution is respectively 65.8 ppb, 16.3 ppb, and 12.6 ppb.

Midwest. The daily mean concentration of total CO over the Midwest region is 258.7 ppb. The North American, Asian, and European CO contribution to the region is 175.3 ppb, 13.9 ppb, and 13.7 ppb respectively. Carbon monoxide concentrations in the Midwest are greater than in the Pacific Coast, due to flux in from the west and large CO emissions from industrial and urban centers in the region like Chicago, Gary, and Detroit.

Mountain West. A strong seasonal trend in the total CO concentrations shows up in the Mountain West, with a summer minimum, similar to the global transport trend. The mean daily total CO concentration is 133.7 ppb. The mean daily contribution of North America, Asia, and Europe is 54.9 ppb, 14.7 ppb, and 9.2 ppb respectively. In mid March of 2001 and 2002, Asian CO concentrations are half of North American CO concentrations (figures 5e and 6e).

Gulf Coast. The Gulf Coast region sees some seasonal variation, but it is not as obvious as in the Mountain West region. The mean daily total CO concentration is 200.0 ppb. The mean daily contribution of CO from North America, Asia and Europe is 119.5 ppb, 12.2 ppb, and 9.3 ppb respectively. The Gulf Coast is host to the port city of New Orleans and the region is ripe with oil exploration and industrial activity (figure 5f and 6f), which explains in part the elevated CO levels in the region.

Northeast. Carbon monoxide concentrations over North America reach their greatest levels over the Northeast, due mostly to large carbon monoxide emissions from fossil fuel use in this heavily populated region. The mean daily total CO concentration is 356.4 ppb. This average is about 100 ppb greater than the Midwest and more than 200 ppb greater than the Mountain West. The North American, Asian, and European mean daily influence is 272.3 ppb, 13.7 ppb, and 14.2 ppb respectively. Asian contribution in the Northeast is less than its contribution on the west coast by about 2-3 ppb. European CO concentrations, however, are greater over the Northeast than elsewhere in North America.

Mid-Atlantic. Like the Northeast, the Mid-Atlantic experiences elevated CO concentrations due to large populations and fossil fuel use. Mean daily total CO concentration is 288.0 ppb. In terms of North

American, Asian, and European contributions, the concentrations are, respectively, 205.1 ppb, 13.3 ppb, and 11.8 ppb.

Southeast. The Southeast region, as shown in figures 5i and 6i, has highly variable daily CO concentrations. The mean daily total is 249.8 ppb. The North American contribution is 168.3 ppb. Similarly, the Asian contribution is 12.3 ppb. European mean daily CO contribution is 9.9 ppb.

Asian and European Impacts on North American CO

Because it is difficult to discern when foreign contributions occur, we plot Asian + European CO on the y-axis with the total CO concentrations on the x-axis (figures 7 and 8). This approach follows *Fiore et al* [2002] which plotted Asian and European ozone against total ozone. The study found that anthropogenic emissions outside North America enhance ozone levels and impact EPA air quality level attainment. For this study, we incorporate the entire population of CO (no averaging) in order to identify transport patterns of by looking at more data points to see foreign concentrations as a function of total CO.

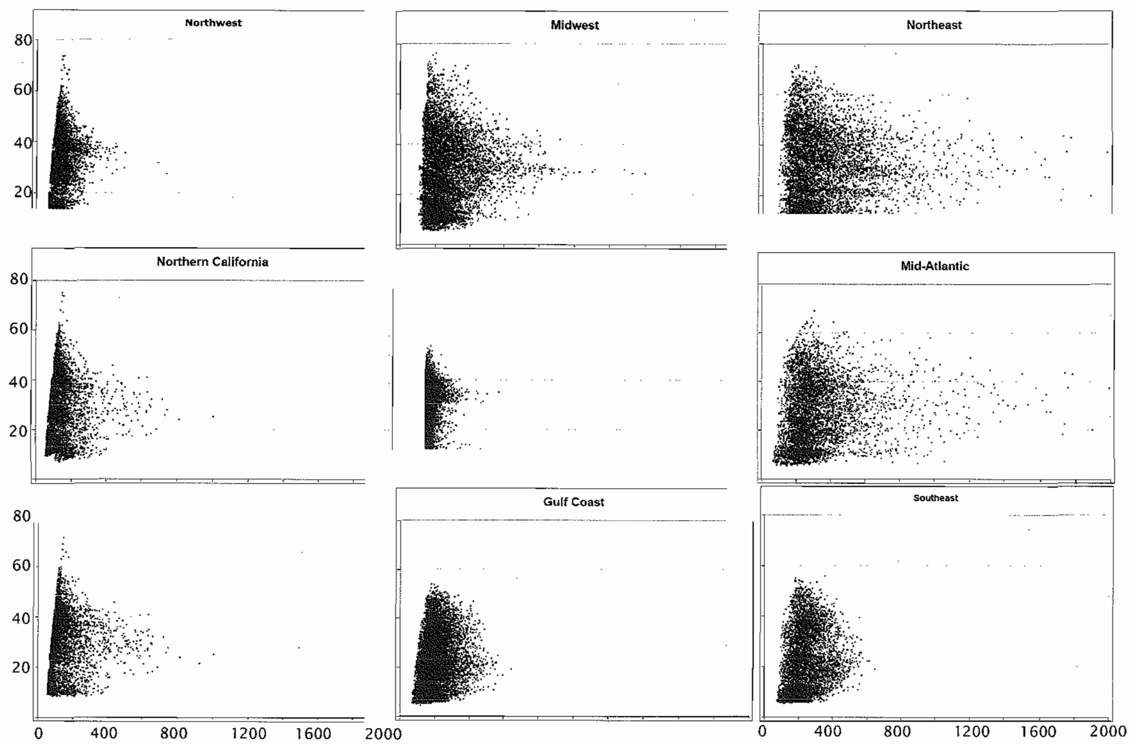


Figure 7a-i: Daily Asian and European CO vs. Total CO over North America in 2001

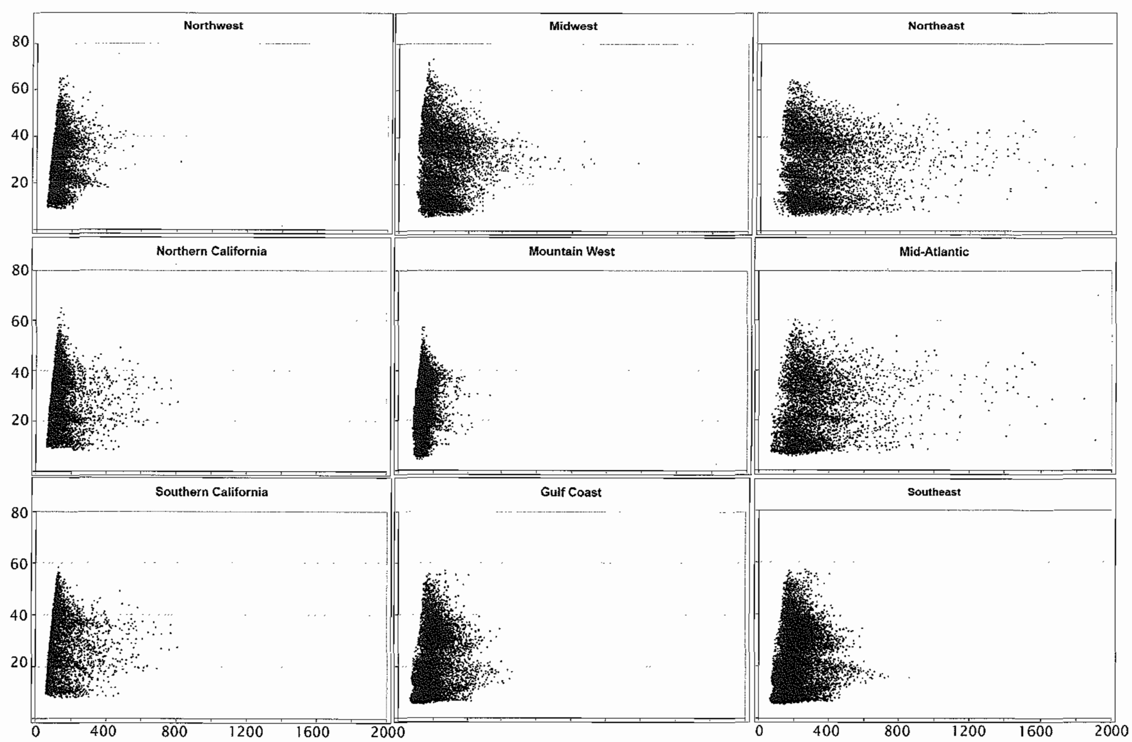


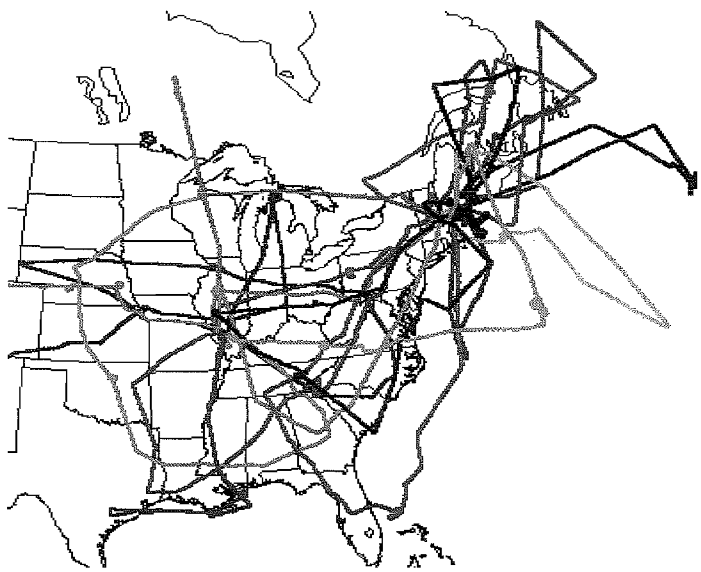
Figure 8a-i: Daily Asian and European CO vs. Total CO over North America in 2002

Figures 7a-i and 8a-i indicate that highest Asian and European CO concentrations occur on days with overall low to moderate total CO concentrations. The low tail of the CO distribution (below 200 ppb) is associated with large enhancements from Asia and Europe, whereas the rest of the distribution is associated with low to moderate Asian and European influence. This means that regions with consistently low to moderate CO levels, like the Northwest and the Mountain West, see a greater influence of Asian and European CO concentrations. Conversely, the large concentrations of CO over the Northeast, Mid-Atlantic, and the Midwest do not feel much of the foreign influence.

While standards for CO have "largely been attained" [*Holmes et al,* 2004], realizing how CO is transported helps to understand how air masses in general are transported and may help in policy making and regulating for other pollutant attainment levels, like ozone [*Fiore et al*, 2002]. We attribute elevated Asian and European influences occurring with low total CO events to polluted air masses originating in Asia and Europe subsiding from the free troposphere to the surface.

V. Transport Pathways of Carbon Monoxide over the Eastern United States

From our comprehension of regional CO in North America, we focus on the eastern United States due to the availability of observations from the INTEX-NA experiment. More specifically, we compare monthly, daily, and hourly MOZART simulations to INTEX-NA observations of CO taken from the NASA DC-8 flights taken the summer of 2004 (figure 9).



NASA DC-8 flight tracks

Figure 9: NASA DC-8 flight tracks in coordination with INTEX-NA, Summer 2004

Among the data collected by the NASA DC-8 aircraft, which were numerous, we analyze carbon monoxide concentrations and observations of CO plumes. NASA DC-8 measured CO concentrations at various heights in the atmosphere throughout the summer of 2004. Individual flight tracks for a given day are shown in a different color. Each flight day consisted of sampling heavily and recording observations of polluted plumes. With these observations, we are able to evaluate the ability of MOZART to simulate regional CO over the eastern United States.

Model evaluation. Figure 10 illustrates the vertical profile of modeled and observed CO. CO from methane, North America, Africa, and Asia are plotted with total modeled CO (red) and total observed CO (black). MOZART agrees well with the observations at higher altitudes, but at the surface, MOZART overestimates carbon monoxide. Given a similar increase at the surface of North American CO concentrations (blue), we attribute MOZART's overestimation of CO at the surface to an overestimation in North American CO emissions. This

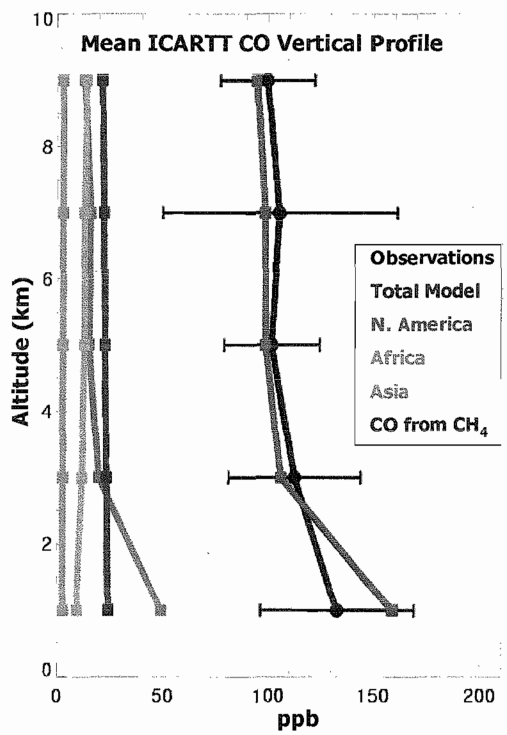


Figure 10: Vertical Profile of Modeled and Observed CO from July 2004

preliminary assessment of MOZART helps with our interpretation of further analysis.

Global North American CO Extent. Figure 11 maps model estimates of global CO concentrations attributable to North American emissions at the surface for July 2004. The main source regions are in the Eastern United States, the greater Los Angeles area, and in Alaska. The high CO concentrations in Alaska and Canada are the result of extensive forest fires during the summer of 2004. North American CO emissions above 10 ppb are carried eastward to western Europe and Greenland via the strong Westerlies aloft. Weaker surface winds carry atmospheric pollutants and other constituents north and eastward.

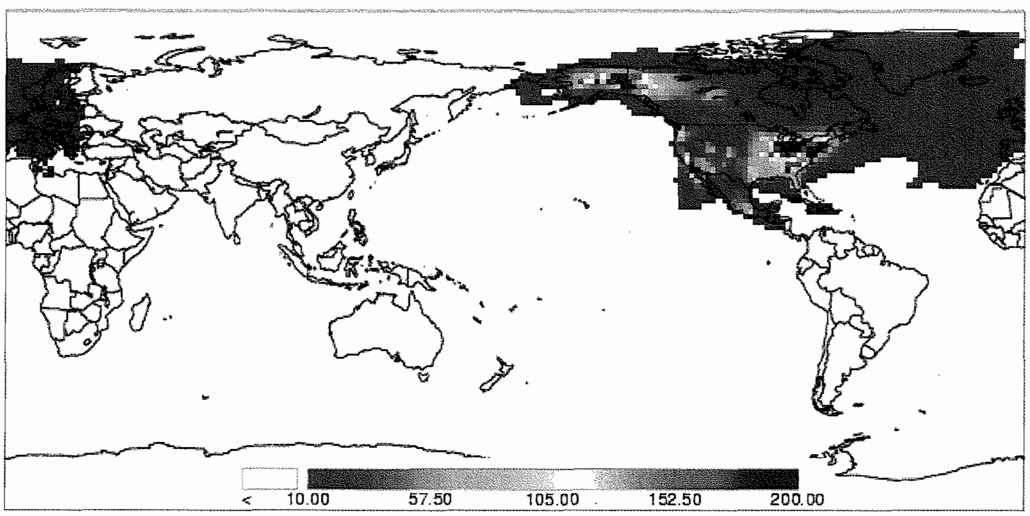


Figure 11: Mean Surface North American Carbon Monoxide Concentrations, July 2004 (ppb)

Global Asian CO Extent. Figure 12 maps the mean Asian surface CO concentrations of July 2004. The main source regions of Asian CO as modeled by MOZART are in India and China, the most populous Asian nations and largest Asian fossil fuel emitters [*Bey et al,* 2001]. It is expected that the rapid industrialization of eastern Asia will have important impacts and changes on not only carbon monoxide concentrations but also atmospheric chemistry in the future [*Bey et al*, 2001 and *Prather et al,* 2001]. Asian CO concentrations in excess of 10 ppb are transported as far as North America, Siberia, and Eastern Europe, with observed fossil fuel and biomass burning signatures [*Bey et al*, 2001]. The Intertropical Convergence Zone (ITCZ) prevents transport of CO southward across the equator. This mechanism also prevents African CO from being directly transported northward across the equator.

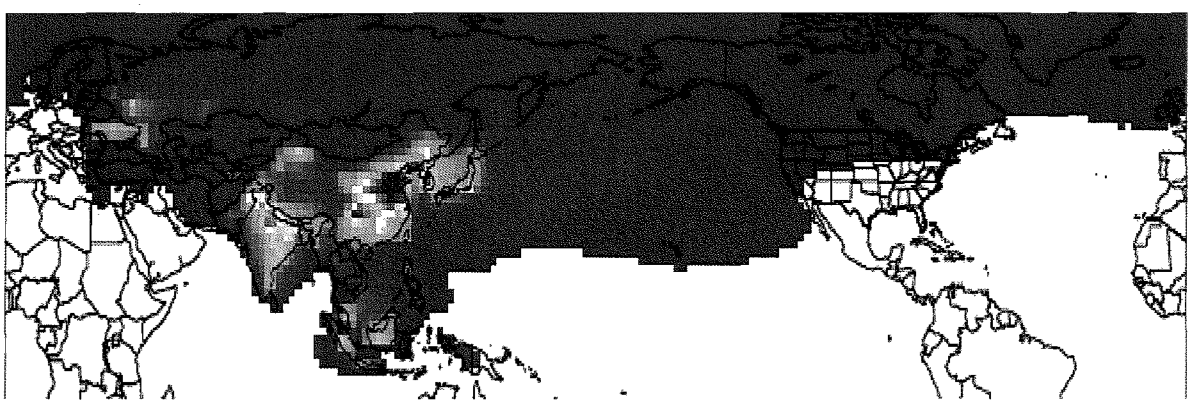


Figure 12: Mean Surface Asian Carbon Monoxide Concentrations, July 2004 (ppb)

Global African CO Extent. African CO concentrations extend across the Atlantic Ocean to South America and a little reaches North America (figure 13). The largest source regions of CO are in the equatorial region and in South Africa. Biomass burning is the main source of CO concentrations over equatorial Africa. The summer is the traditional burning season in Africa. Because biomass burning emissions occur in the tropics, the lifetime for African CO is shorter than Asian or European CO due to rapid oxidation with high concentrations of OH in low latitudes [*Bey et al,* 2001]. The ITCZ prevents hemispheric transport northward; however residual CO does contribute to eastern United States CO levels.

Figure 13: Mean Surface African Carbon Monoxide Concentrations, July 2004 (ppb)

After looking at global concentrations of CO, we map such concentrations over the eastern United States for comparison with NASA

DC-8 measurements to understand how North American, Asian, and African source regions impact the regional scale.

Origins of CO over the Eastern United States

North American. North American mean carbon monoxide concentrations over the eastern United States in July 2004 are greatest at the surface, near the source region where fossil fuel emissions are very high. Large concentrations also occur at higher latitudes, due in part because of northward and eastward surface winds (shown in white arrows in figure 14b). Low OH concentrations at high latitudes also contribute to greater carbon monoxide concentrations at these higher latitudes. At 400mb in the troposphere (figure 14a), North American CO concentrations are smaller, with the greatest concentrations centered over and off the coast of New York City.

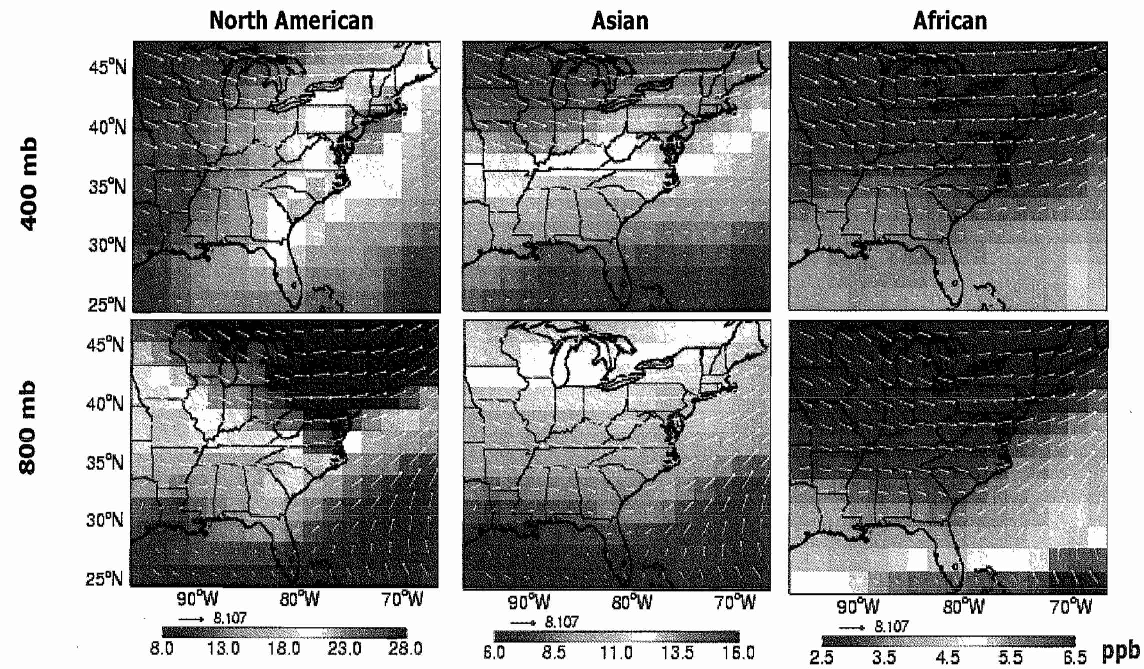


Figure 14: Mean North American, Asian, and African CO over the eastern United States, July 2004 (ppb)

Asian. Asian carbon monoxide concentrations over the eastern United States are greatest higher in the atmosphere and over the north central Midwestern U.S. (figure 14c). The highest magnitude of Asian CO over the eastern United States is 16 ppb, whereas the greatest magnitude of North American CO for the same region is 26 ppb. The greater Asian concentrations aloft suggest transport westward via the strong Westerly winds aloft. Pollutants transported in the free troposphere (2-12 km above the surface) will be carried further and faster than pollutants transported in the boundary layer (surface to 2 km above) [*Fiore et al,* 2003]. Asian CO transport events occur more often above the boundary layer [*Yienger et al*, 2000; *Wild et al*, 2004]. The surface Asian CO concentrations (figure 14d) are minimal, with a greater concentration at higher latitudes.

African. The presence of African carbon monoxide concentrations over the eastern United States is small, ranging from 2.5 to 6.5 ppb (figure 14e, f). African CO is greatest at the surface at southern latitudes (figure 14f). African CO concentrations aloft are minimal but still greatest at lower latitudes (figure 14e). African CO concentrations are the residual effect of heavy biomass burning in Africa during July of 2004. African carbon monoxide concentrations are greater at the surface because, unlike in Asia, the tropics and subtropics are not dominated by uplift. CO from Africa reaches North America not by a direct transport mechanism, but rather by residual concentrations that are circulated clockwise by the Bermuda high from the tropics, which streams air northward across the Atlantic and Gulf of Mexico towards the southeastern part of the U.S. and sometimes up to New England before curving eastward out into the Northern Atlantic.

Daily Variability of Eastern U.S. Summer CO Fluxes and Concentrations

Characteristics of these transport patterns become clearer by examining fluxes (Tg/day) into and out of the eastern United States region.

Flux Variability. Fluxes are used to quantify transport flow rates in terms of the amount of carbon monoxide (in Tg) that passes through a unit area per unit of time, in this case, day. The eastern United States is the study area, bounded at 97°W on the west and 67°W on the east. Flux values are modeled as the necessary observations along the boundaries do not exist. Flux into the region is defined as positive when CO is transported past 97°W from the west; flux out is defined as positive when transport CO passes 67°W also from the west (figure 15). By modeling CO fluxes, we estimate daily CO burdens and the contribution of foreign CO fluxes that are transported via winds.

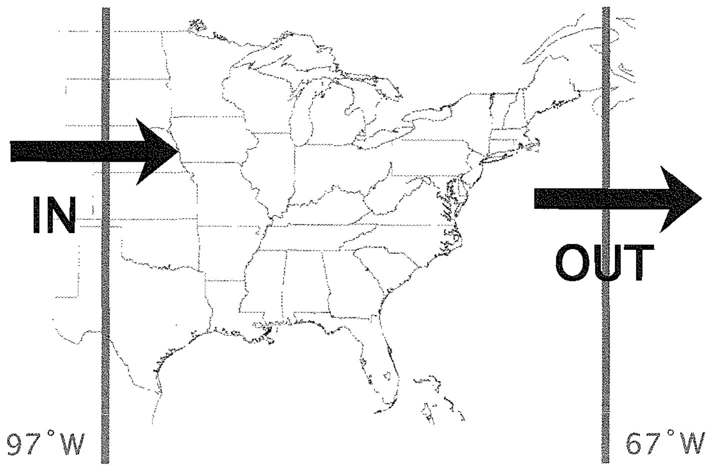


Figure 15: Schematic of Flux in and out as defined over the eastern United States

Plots of modeled fluxes show a high amount of daily variability from July to mid-August of 2004, shown by the many peaks and troughs and the varying magnitudes of each. The total CO flux (black) has the greatest variability, influenced most by North American and Asian CO fluxes. There is also a temporal lapse between the flux in (figure 16a) and the flux out (figure 16b), with large peaks in fluxes out following large peaks in fluxes into the region.

Fluxes into the region are smaller than the fluxes out of the region, due to greater CO burdens (Tg) from large eastern United States CO emissions. Asian fluxes into the eastern United States at 97°W is significant and for most of the year and larger than North American flux in.

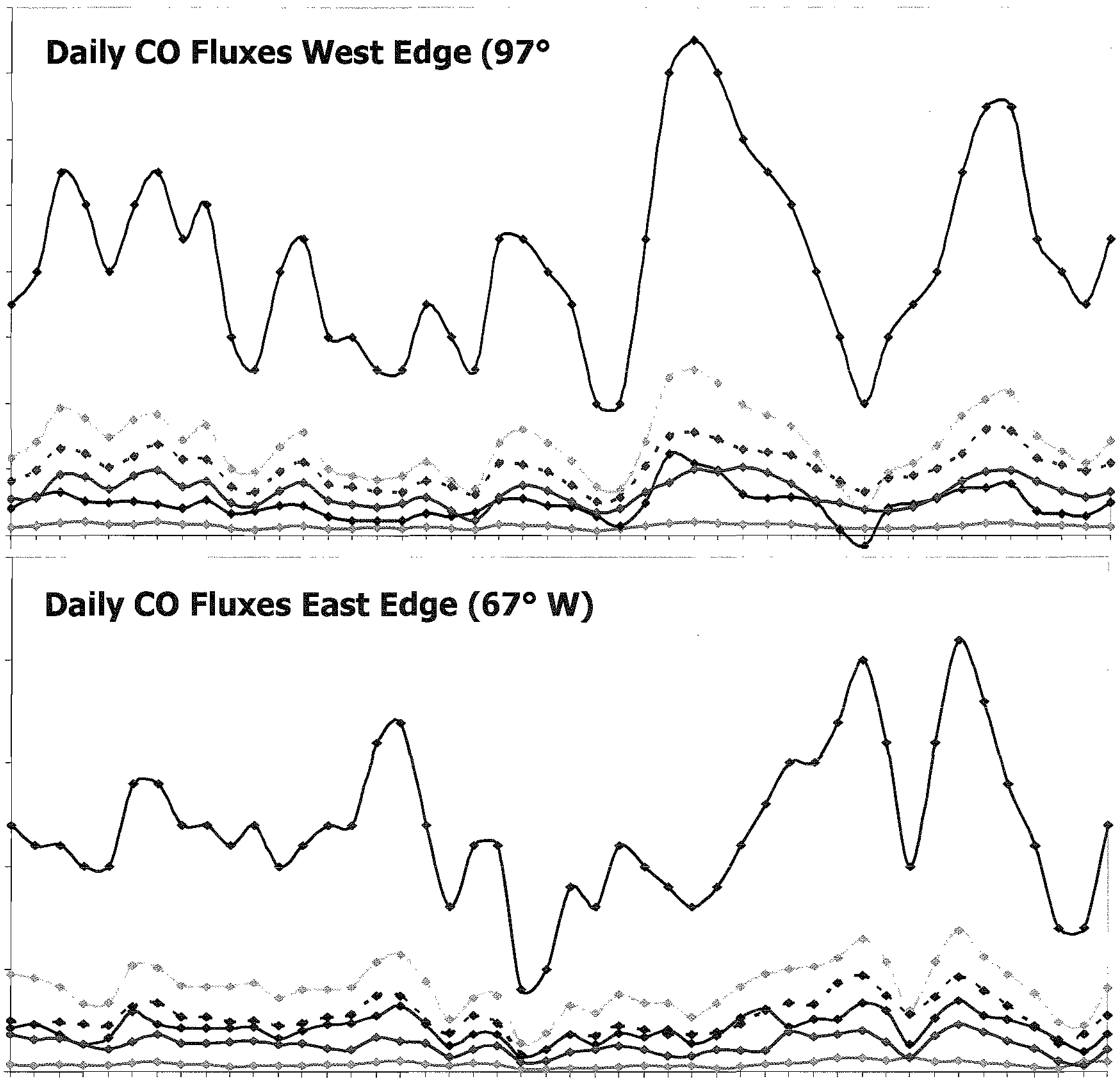


Figure 16a-b: CO Flux In (a) and out (b) for Summer 2004 (Tg/day) CO fluxes shown: Total (black), African (light green), North American (blue), All regions (yellow), Asian (red), from methane (black dashed). July- August 15, 2004

Asian CO fluxes into the region and out of the region are similar, whereas North American flux out is greater than the North American flux in. This is easily explained by the large amount of carbon monoxide emissions in the eastern United States, increasing the atmospheric CO burden. African CO flux in and out of the eastern United States shows little to no daily variability through the time series.

Modeled and Observed Daily CO. Peak flux events on August 2 and July 18 of 2004 coincided with NASA DC-8 aircraft observations and motivated a closer analysis of MOZART-simulated CO.

August 2, 2004. Analysis of August 2, 2004 simulation with observations reaffirms some of the broader patterns already illustrated. Carbon monoxide concentrations derived by MOZART (red) and observed (black) are plotted hourly with Asian CO, North American CO, African CO, methane CO, and altitude also identified (figure 17).

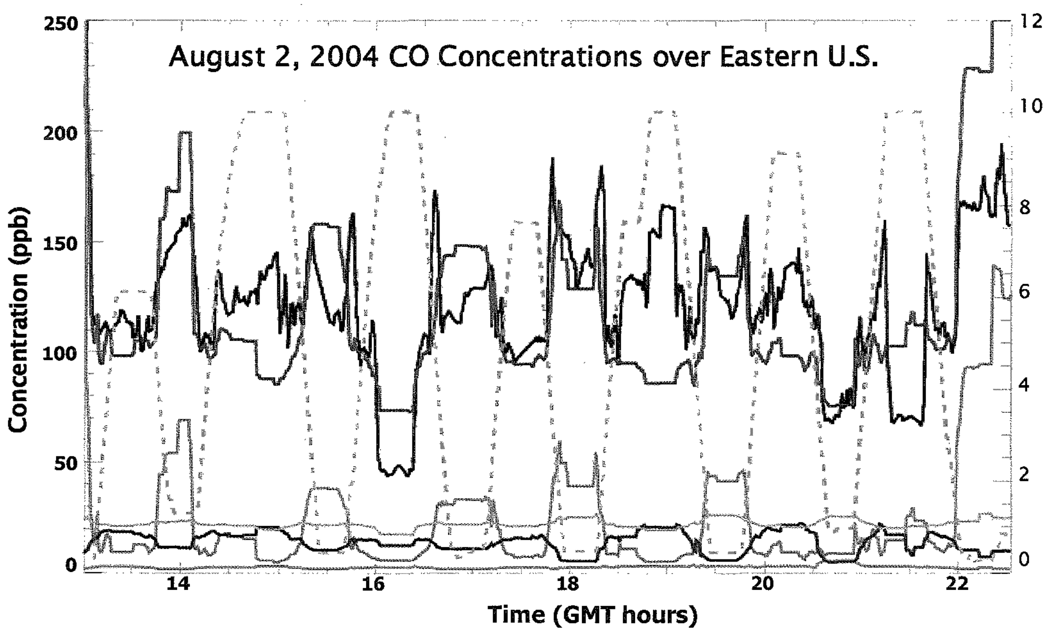


Figure 17: August 2, 2004 Hourly CO Concentrations over the Eastern U.S. CO (in ppb) is plotted: Observations (black), Total modeled (red), North American (blue), African (green), Asian (purple), from methane (gold), altitude (gray) in km and uses right y-axis

Some key patterns arise within total and regional carbon monoxide concentrations. MOZART accurately captures the general hourly trend but misses the magnitude of some plumes. The total modeled carbon monoxide concentrations are driven mainly by North American CO patterns; however varying plane altitudes (gray) also impact the magnitude of North American and Asian CO. When the plane's path is high in the atmosphere, greater Asian CO concentrations are modeled. Likewise, when the plane is lower in the atmosphere, modeled North American CO concentrations are greater.

Asian and North American carbon monoxide concentrations are plotted at two different heights in the atmosphere (figure 18) on August 2, 2004. At 800 mb, near the surface, North American CO concentrations are greatest at higher latitudes, following OH concentration characteristics and winds (figure 18a). Summertime cyclones over North America are displaced further northeastward which, in addition to OH characteristics, explains the concentration of North American CO at higher latitudes and off the northeast shore [*Li et al*, 2005].

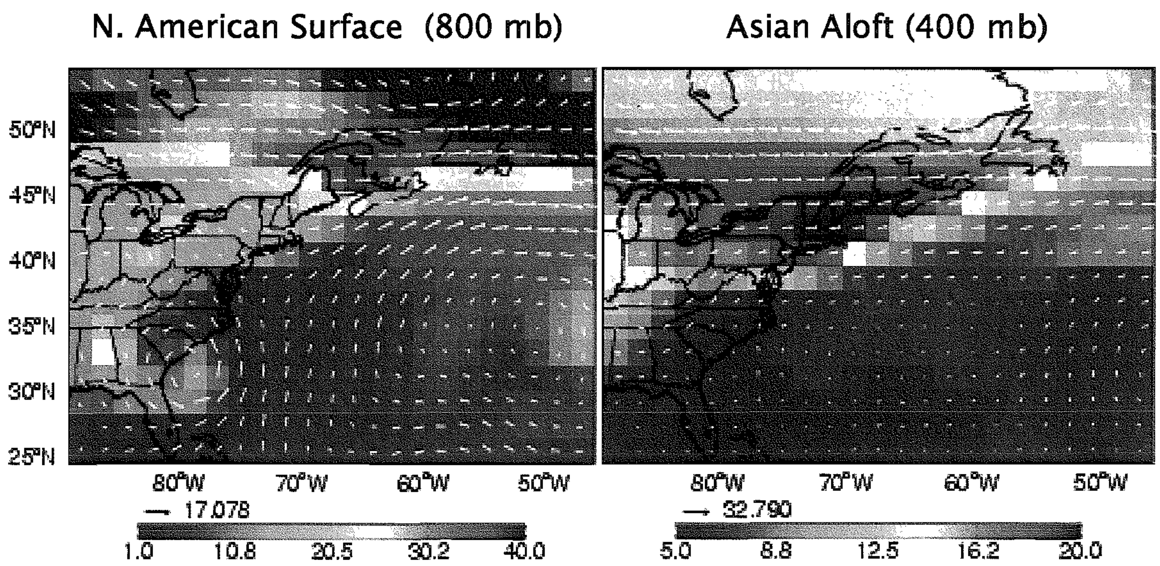


Figure 18: North American surface CO (800 mb) and Asian aloft CO (400 mb) on August 2, 2004

At 400 mb, higher in the troposphere where the winds are stronger, Asian CO concentrations are greatest also at higher latitudes (figure 18b).

July 18, 2004. Like August 2, simulated hourly carbon monoxide concentrations are plotted with measured CO (figure 19). As seen on August 2, total modeled and observed carbon monoxide agree temporally, but July 18 illustrates the inability of MOZART to capture individual CO plume events, like the observed biomass burning plume at 19:00 Greenwich Mean Time (GMT). This plume is attributed to the forest fires of western and northern North America that occurred that summer. While fossil fuel emissions are relatively constant throughout the year and

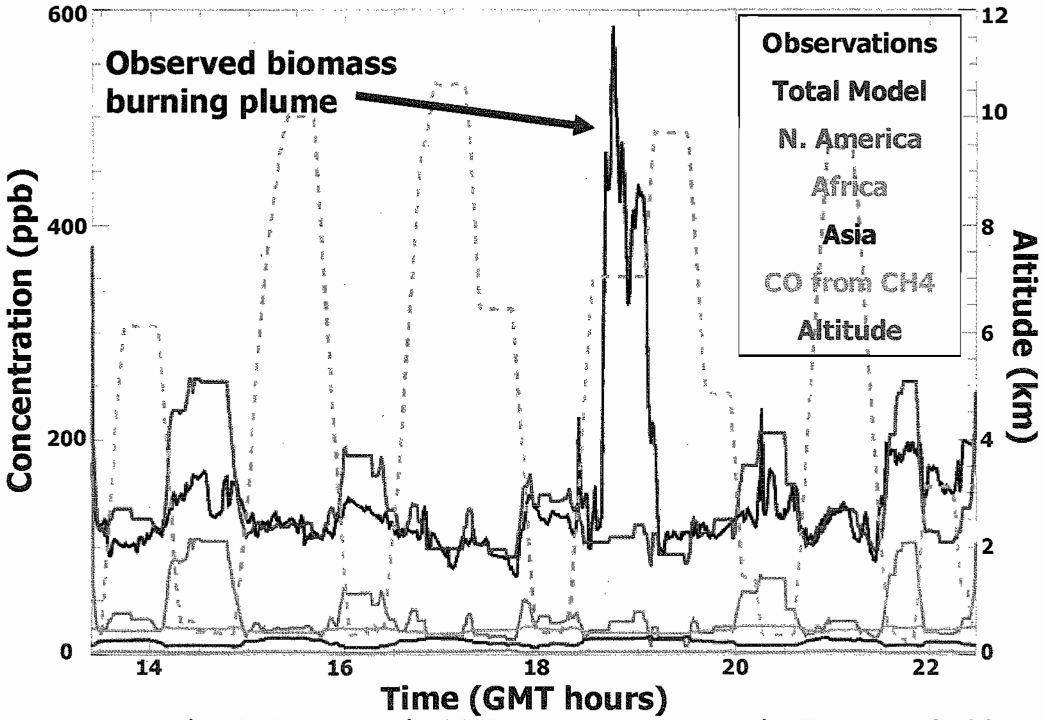


Figure 19: July 18, 2004 Hourly CO Concentrations over the Eastern U.S. CO

control large scale CO concentrations, biomass burning emissions are more temporally varied and impact regional CO *[Holloway et al*, 2000]. In general, the MOZART-simulated CO is greater than observed CO, but this is attributed to an overestimation of North American CO emissions, as was described earlier.

Specific Transport Pathways. While MOZART was not able to recreate some observed plumes and magnitudes, it does capture temporal variability and the nature of intercontinental transport of carbon monoxide for the eastern United States. Based on concentrations and fluxes, we define three main transport pathways of CO over the eastern United States (figure 20).

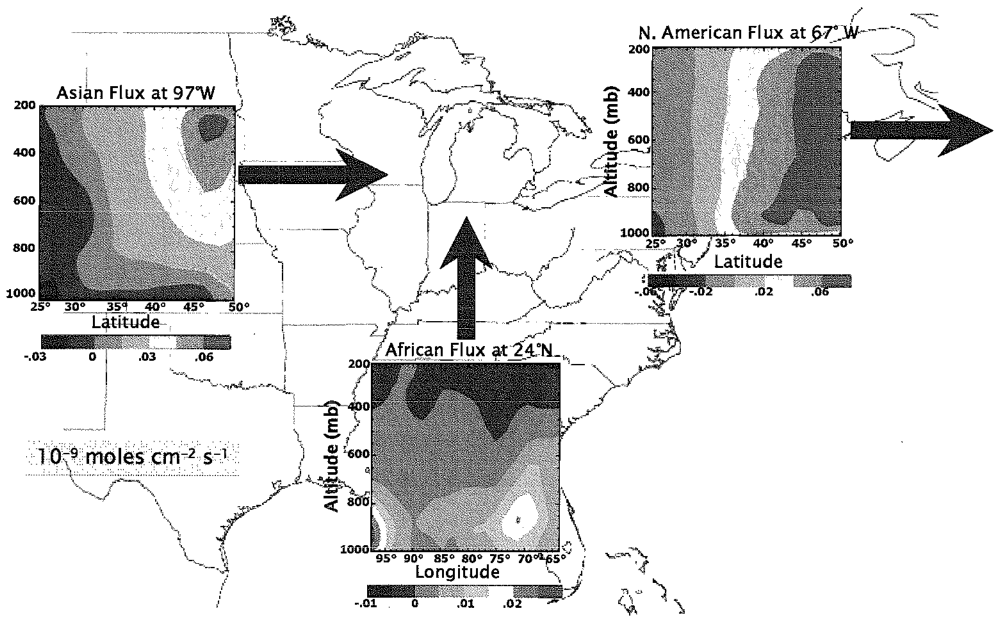


Figure 20: CO Transport Pathways in the eastern U.S.

Asian carbon monoxide fluxes and concentrations are greatest at higher altitudes and latitudes, suggesting transport aloft via the Westerlies. African carbon monoxide fluxes and concentrations are greatest at the surface and at southerly latitudes, indicating the residual effect of summertime biomass burning in Africa. North American carbon monoxide fluxes and concentrations are greatest at all levels, at higher latitudes, transporting eastward out of the region.

VI. Conclusion

Nine regions over North America, including an in-depth study of the eastern United States, were studied to understand the impact of intercontinental transport of carbon monoxide at the regional level. By incorporating multiple methods in a qualitative overview, the intercontinental transport of carbon monoxide is defined over North America. Based on a global model analysis at the regional scale, intercontinental transport of CO from Asia and Europe is highest on days with an overall low to moderate total CO concentrations. Mean daily Asian carbon monoxide concentrations are greatest on the Pacific Coast, while overall total daily carbon monoxide levels are greatest on the Atlantic Coast.

In the eastern United State, modeled and observed carbon monoxide concentrations and modeled fluxes show evidence of three main transport pathways for summertime carbon monoxide. North American carbon monoxide concentrations in the eastern United States are greatest at the surface and higher latitudes. These concentrations are transported northward and eastward toward the north Atlantic.

Recognizing the regional CO influence and the pathways of transport is a step towards understanding how polluted air masses evolve and impact regional air quality. By understanding how carbon monoxide is transported, we can understand global transport of other pollutants, like ozone, as well. Transport has important policy implications as Asian CO emissions are not projected to slow [*Fiore et al*, 2002 and *Prather et al,* 2001]. While policy makers push to reduce local and regional emissions, it is necessary to know the local effect of foreign sources and its impact on regional air quality especially as global communities are working together to address intercontinental transport of air pollution.

References

Allen, D.J., et al. "Transport-induced interannual variability of carbon monoxide determined using a chemistry and transport model." *Journal of Geophysical Research* 101 (1996).

Berntsen T.K. and I.S.A. Isakson. "A global three-dimensional chemical transport model for the troposphere, 1, Model description and CO and ozone results." *Journal of Geophysical Research* 102 (1997).

Bey, I., et al. "Background ozone over the United States in summer: Origin, trend, and contribution to pollution episodes." *Journal of Geophysical Research* 107.D15 (2002).

Bey, I., et al. "Asian chemical outflow to the Pacific in spring: Origins, pathways, and budgets." *Journal of Geophysical Research* 106.D19 (2001).

Fehsenfeld, F.C., et al. "International Consortium for Atmospheric Research on Transport and Transformation (ICARTT): North America to Europe—Overview of the 2004 summer field study." *Journal of Geophysical Research* 111.D23 (2006).

Fiore, A., et al. "A Global Perspective on Air Quality: Intercontinental Transport and Linkages with Climate." *EM* 13-22: (2003).

Holloway, T., et al. "Global distribution of carbon monoxide." *Journal of Geophysical Research* 105.D10 (2000).

Holmes, K.J., et al. "Why Carbon Monoxide Still Matters." *Environmental Science and Technology* 289A-295A: (2004).

Horowitz, L., et al. "A global simulation of tropospheric ozone and related tracers: Description and evaluation of MOZART, version 2." *Journal of Geophysical Research* 108.D8 (2003).

Jacob, D. *Introduction to Atmospheric Chemistry.* Princeton: Princeton University Press, 1999.

Kanakidou, et al. "3-D global simulations of tropospheric CO distributions – results of the GIM/IGAC intercomparison 1997 exercise." *Chemosphere: Global Change Science* 1 (1999).

Kiley, C., et al. "An intercomparison and evaluation of aircraft-derived and simulated CO from seven chemical transport models during the TRACE-P experiment." *Journal of Geophysical Research* 108.D21 (2003).

Li, Q., et al. "North American pollution outflow and the trapping of convectively lifted pollution by upper-level anticyclone." *Journal of Geophysical Research* 110.D10301 (2005).

Muller, J.-F. and G. Brasseur. "IMAGES: A three-dimensional chemical transport model of the global troposphere." *Journal of Geophysical Research* 100 (1995).

Novelli, P., et al. "Recent Changes in Atmospheric Carbon Monoxide." *Science* 263.5015 (1994).

Pinto, J.P., et al. "A general circulation model study of atmospheric carbon monoxide." *Journal of Geophysical Research* 88 (1983).

Prather, M. and D. Ehhalt. "Atmospheric chemistry and greenhouse gases." *Climate Change: The Scientific Basis: The IPCC Working Group 1 Third Assessment Report.* New York: Cambridge UP, 2001.

Saylor, R.D. and L.K. Peters. "The global numerical simulation of the distribution of CO in the troposphere." *Air Pollution Modeling and Its Applications VIII.* Ed. H. van Dop and D.G. Steyn. New York: 1991.

Shidell, D.T., et al. "Multimodel simulations of carbon monoxide: Comparison with observations and projected near-future changes." *Journal of Geophysical Research* 111.D19306 (2006)

Singh, H., et al. "Overview of the summer 2004 Intercontinental Chemical Transport Experiment - North America (INTEX-NA)." *Journal of Geophysical Research* 111.D24 (2006).

Wang, Y., et al. "Global simulation of tropospheric O_3-NO_x-hydrocarbon chemistry." *Journal of Geophysical Research* 103 (1998).

Wild, O., et al. "Trans-Eurasian transport of ozone and its precursors." *Journal of Geophysical Research* 109.D11302 (2004).

Wilkening, K.E., et al. "Trans-Pacific Air Pollution." *Science* 290, 65 (2000).

Yienger, J., et al. "The episodic nature of air pollution transport from Asia to North America." *Journal of Geophysical Research* 105.D22 (2000).