TRANSPORT OF AIR POLLUTION FROM ASIA TO NORTH AMERICA

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INTRODUCTION

Recognizing a potential for trans-Pacific transport, scientists are beginning to look for an Asian pollution signal over North America (NA). Kritz et al. [1990] previously found that Asian boundary layer air can be transported to the upper troposphere over California in two to four days. More recently, Jaffe et al., [1999] published evidence of Asian air pollution transported to the surface of North America during spring. The year following Jaffe's 1997 observations, a strong April Asian dust storm carried a visible plume of dust to North America [Husar, 1998]. There is growing awareness of the need to include an intercontinental perspective when analyzing North American chemical and aerosol data. Indeed, evidence of trans Pacific transport continues to mount; Jaffe's group has once again detected in the spring of 1999 significant plumes of Asian pollution off of Washington state [Dan Jaffe, private communication].

In this study we employ the Geophysical Fluid Dynamics Laboratory (GFDL) Global Chemistry Transport Model (GCTM) for CO and O_3 to specifically address the episodic nature of trans-Pacific pollution. The model's resolution (265 km x 265 km) is sufficient to produce synoptic-scale tracer fluctuations similar in magnitude and nature to those observed in the real atmosphere [see Levy and Moxim, 1989; Moxim, 1990; Moxim et al., 1996]. We address questions such as how well is the Asian pollution signal over North America represented by a mean value? How frequent are strong transport events (i.e., like those observed by Jaffe et al. [1999] at CPO) expected during and outside of spring, and how does this frequency differ with location and altitude?

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MODEL DESCRIPTION

GCTM

The Geophysical Fluid Dynamics Laboratory (GFDL) Global Chemical Transport Model (GCTM) has a horizontal resolution of ~265km [2.4°x2.4° in the tropics and 3°-3.5°x2.4 in midlatitudes], 11 sigma levels in the vertical at standard pressures of 10, 38, 65, 110, 190, 315, 500, 685, 835, 940, and 990 mb and is driven by 12 months of 6-hour time-averaged winds, temperatures, and precipitation fields from a general circulation model [Manabe et al., 1974]. The transport portion of the GCTM employees a numerical scheme which is 2^{nd} order in the horizontal and 4th order in the vertical for resolved (grid-scale) advection [see Section 3. of Mahlman and Moxim, 1978] and includes diffusion-based parameterizations for horizontal sub-grid scale transport and for vertical sub-grid scale transport due to dry and moist convection throughout the troposphere and shear dependent mixing in the boundary layer [Kasibhatla et al., 1996; Levy et al., 1999 and references therein for details].

CO and O_3 Simulations

This work relies on the analysis of global CO and O_3 simulations that have been described and extensively evaluated previously [Levy et al., 1997; Klonecki and Levy, 1997; Yienger et al., 1999; Holloway et al., 1999]. The GCTM simulation of CO is designed for 1990 conditions that include emissions from fossil fuel [300 Tg CO/yr], biomass burning [748 Tg CO/yr], biogenic hydrocarbon oxidation [683 Tg CO/yr] and CH_4 oxidation [758 Tg CO/yr]. The only CO destruction pathway is OH oxidation based on pre-calculated monthly-mean 3D OH fields [Spivakovsky et al., 1990] that have been scaled by 1.15 to give a CH_3CCl_3 global lifetime of 4.8 years..

The GCTM simulation of ozone is a one tracer experiment with four component sources and sinks. At the tropospheric boundaries, ozone is carried down from the stratosphere by Stratosphere-Troposphere Exchange [STE] and destroyed at the surface via dry deposition. In the polluted BL, net ozone production is computed with a NO_x-dependent proportionality constant that relates NO2 oxidation to HNO_3 to net ozone production. In the free troposphere and clean BL, instantaneous production and destruction are based on diurnal average steady-state solutions to a CH_4-CO/Acetone-H_2O-NO_x photochemical box model applied over land and sea for the global range of latitudes and model levels and for every month. This battery of solutions for the tropospheric ranges of CO/Acetone, NO_x, O_3 and H_2O comprise an off-line table that is accessed every time-step in every grid-box with the appropriate values of NO_x and CO from previous simulations (Levy et al., 1999; Holloway et al., 1999), O_3 from the current simulation, and monthly averaged water vapor (Oort, 1983; Soden and Bretherton, 1996). Both a detailed discussion of this method and an extensive comparison to observations can be found in Levy et al. [1997], Klonecki, [1998], and Yienger et al. [1999].

Definition of Asian and North American CO and O_3 Tracers

To isolate pollution from Asia, we made three separate 2-year CO simulations: one with full global emissions, one without surface emissions from Asia (defined to include South, Southeast, and East Asia), and one without surface emissions from North and Central America. In the last two cases, biogenic hydrocarbon oxidation is considered an instantaneous surface source of CO and is removed along with fossil fuel and biomass

burning emissions of CO. The first year of the run is used to equilibrate the model and the second year used in analysis. We define the contribution of either Asia or North America to total CO (tracers we refer to as "Asian CO" and "North American CO") as the difference between the full emission simulation and the respective simulations without the local emissions. For ozone, analogous pairs of simulations were first run and differenced for NO_x and then O_3 simulations were run with the appropriate CO and NO_x fields to produce both 1990 and future "Asian O_3" tracers. See Levy et al. [1999] for a description of the NO_x sources used in the 1990 base-year simulations, and Yienger et al. [1999] for the methodology used to scale these global sources to 2020. Both 1990 and future-projected NO_x emissions for South, Southeast and East Asia are taken from Van Aardenne et al. [1998].

ASIAN CO TRANSPORT TO NORTH AMERICA

Signal Variability Across North America

In Figure 1 we show the time series of Asian CO for each of the three points considered in this study (California, CPO, and Central Canada as shown in Figure 1) for both the BL (940 mbar) and the mid troposphere (500 mbar). In order to illustrate the number of events analogous in strength to those observed at CPO, we include a line at 60 ppb that defines the average amount of Asian CO we estimate to have been present during the real events observed by Jaffe et al., [1999]. In arriving at 60 ppb as the indicator of Asian CO events, we noted that the actual episodes at CPO had a mean difference of ~20 ppb between trajectories segregated as Asian and non-Asian (168 ppb vs. 150 ppb).

Figures 1a-f reveals that the frequency and magnitude of Asian CO episodes typically increase with height and towards the mid-latitudes. In the middle troposphere (500 mb) over CPO, Figure 1b, the springtime Asian signal is stronger than in the BL (940 mb). There are numerous spikes over 60 ppb implying that many significant Asian pollution events pass overhead without being seen at the ground. Throughout the whole spring, such a CO signal occurs 20% of the time, as opposed to only 4% of the time in the BL. Trajectories from the GCTM reveal that flow from Asia is much more common aloft than in the BL. At the ground, flow reaching CPO was more likely to have been diverted from higher latitudes around the Aleutian Low than to have come straight from East Asia. Moving southward to central California the Asian CO signal in the BL is similar to CPO (Figure 1c). There are four distinct 60+ ppb episodes and a few extended periods in March and April that are close. The individual events are not as strong, and as a whole the Asian signal is a little less noisy than at CPO. This may be because the CO was more likely to have been transported to the surface via subsidence than at CPO where events episodically arrived via low level flow (< 700 mbar) all the way across the Pacific (a more detailed description of transport will be presented in the Section 5). Aloft at 500 mbar (Figure 1d) there is a large increase in the Asian CO signal that is even stronger than the relative increase over CPO. Here, Asian CO exceeds 60 ppb in spring nearly 50% of the time, and exceeds 100 ppb in six separate episodes. In contrast to California, the signal in north central Canada is much weaker. The BL signal (Figure 1e) is muted and nearly always representative of the mean enhancement, even in spring. Aloft (Figure 1f) it is much weaker than in the lower latitudes but still significantly enhanced over the BL. About eight separate spring events equal or exceed 60 ppb Asian CO, and their respective back trajectories originate from Asia. Most come from northern China and Japan, although the large spike in early February originates from South East Asia. In this

particular transport event, air containing more than 60 ppb Asian CO was lifted from the south east Asian BL and transported, in 9 days, all the way to north central Canada.

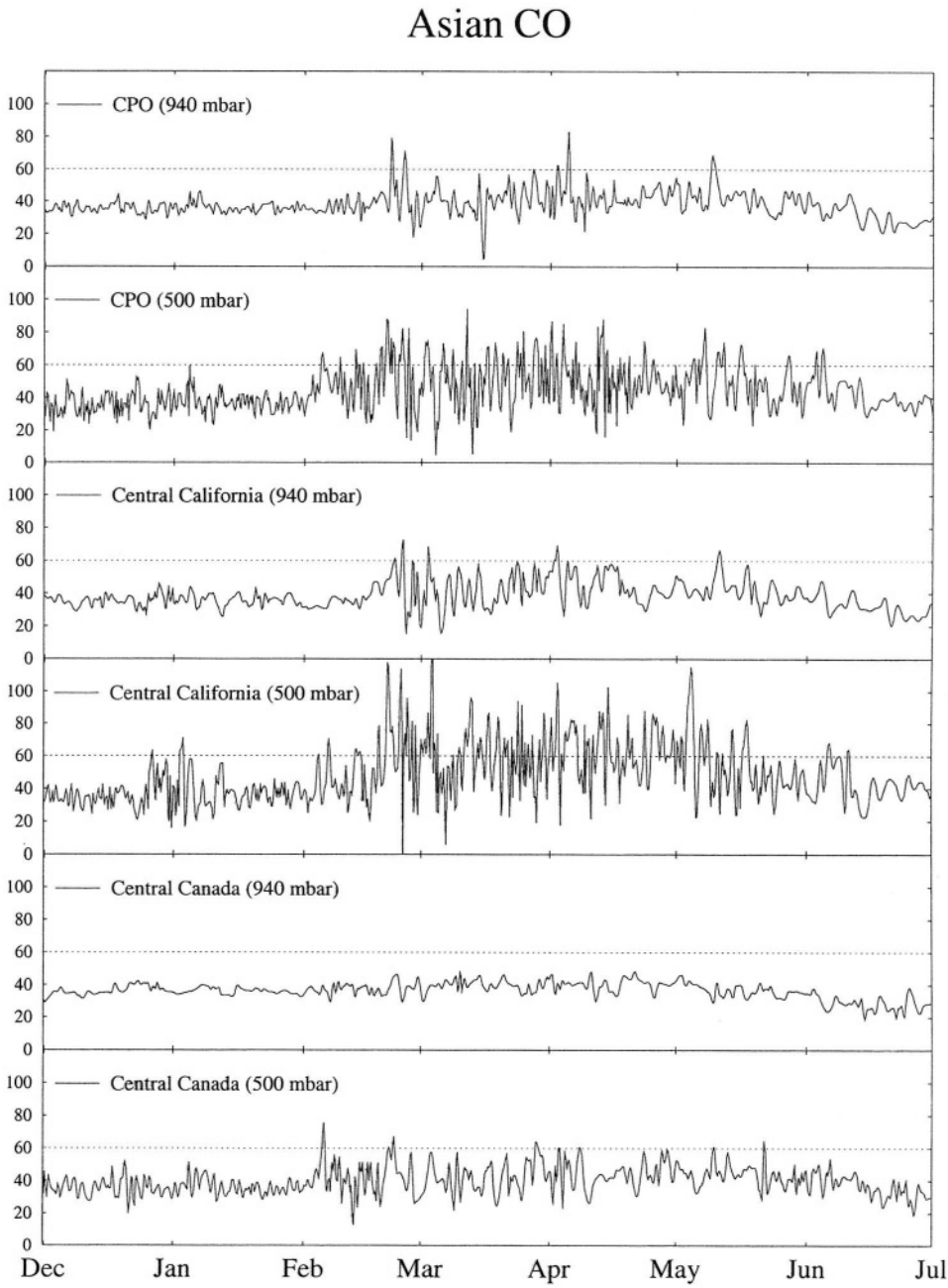

Figure 1. Time series at 6-hour intervals of Asian contribution to CO at all three points at both 940 mbar (BL) and 500 mbar (mid-troposphere). A line at 60 ppb illustrates the amount of Asian CO we estimate to have been present in the pollution parcels observed by Jaffe et al. [1999] at CPO.

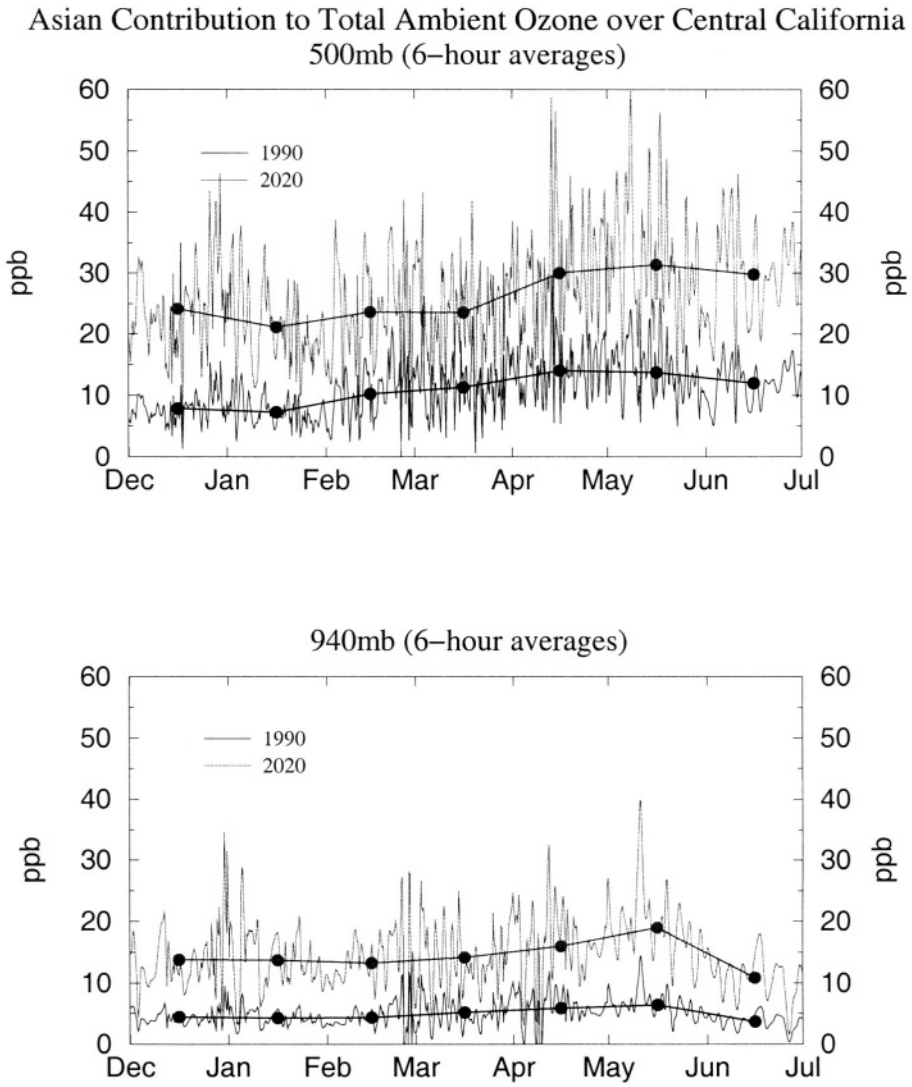

Figure 2. Asian contribution to total ozone offshore of central California at (a) 500 mbar and (b) 990 mbar. In both figures we present the time series for both 1990 emissions and the worst-case-scenario 2020 emission rates.

IMPACT OF ASIA ON OZONE ACROSS NORTH AMERICA

Episodic Enhancements

If there is a significant impact from Asia on US ozone levels, whether it be on surface air quality or on ozone variability in the free troposphere, we believe it probably occurs during episodic events when Asian contributions are unusually high. In Figure 2 we present the time series of Asian O_3 for 1990 in the grid box used to analyze CO directly off the coast of California. On the same figure we also plot the corresponding contributions when Asian emissions are increased under the future scenario to demonstrate the sensitivity of the episodes to increasing emissions. The Asian signals are highly episodic. In each case the strongest events occur in April-May, as opposed to late February-early May for CO, probably due to a lag between the maxima in physical outflow from Asia and net photochemical ozone production. At 500 mbar with 1990 emission levels (Figure 2a), 4 events with greater than 20 ppb of Asian ozone occur in May and two each in March, April and late February. Under the future scenario, extreme springtime events at 500 mbar now approach 60 ppb, and most events greater than 40 ppb have shifted to April and May, periods which have more active photochemistry. This behavior is consistent with previous global 3-D simulations that show that increasing emissions shift the timing of the mid-latitude springtime ozone maximum to later into spring (e.g. Yienger et al., 1999).

In the BL (Figure 2b), at 1990 emission levels a few Asian episodes exceed 10 ppb in spring but never exceed 5 ppb in summer. This suggests that even on an episodic basis, the impact of Asia on ozone this past decade has been small. On the other hand, as Asian NO_x emissions grow to 25 Tg N/yr or more, short-term spikes of Asian ozone in the BL could reach 30-40 ppb in May, and perhaps 10-20 ppb in summer. The 40 ppb May episode seen in Figure 2b accounts for roughly half of the total ozone in that parcel (~80 ppb). From the perspective of air quality accounting, if only three 8-hour 80 ppb episodes will be needed for exceedance, then a few strong trans-Pacific events may aggravate local pollution enough to tip the balance in some areas. This is particularly true in higher altitude urban or suburban areas more exposed to free-tropospheric air. Even if actual future emissions are not so strong, smaller episodes of 20-30 ppb Asian ozone (out of perhaps 60-70 ppb total) would still be significant in the US BL.

CONCLUSIONS

There are a number of implications of these results. For example, those measuring atmospheric chemistry over NA typically have very little information about potential significance of trans-Pacific transport, particularly on short time scales when most measurements are taken. Our results suggests that NA aerosol and chemical data may need to be reconciled with intercontinental transport to explain synoptic-scale variability, particularly for enigmatic high episodes that appear to have no local origin. For regional scale chemical modeling that relies on static or monthly averaged boundary conditions for either Pacific air or descending free tropospheric air over NA, this study provides insight into the potential synoptic-scale consequences of these boundary condition assumptions. Results presented in this paper suggest that regional scale models that pose static chemical boundary conditions for Pacific "background" air may suffer from unexplained variability in tracer time series when compared to observations. Finally, from an ozone policy perspective there could be significantly different consequences of an Asian ozone signal that arrives as a smooth constant background increase, versus one that arrives episodically as a mixture of weak and strong events. Episodes that

contribution 40 ppb of ozone to surface ozone levels in NA due to Asian emissions are certain to exacerbate local pollution events and contribute significantly to the frequency of elevated (80 - 100 ppb) ozone exposures. This is especially of concern in elevated areas more frequently exposed to free tropospheric air.

We invite the reader to the following web site *[http://www.cgrer.uiowa.edu/asiaimpact]* that contains a number of related animations of trans-Pacific transport.

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