Title
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Permalink
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Journal
Geophysical Research Letters, 33(1)

ISSN
0094-8276

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Publication Date
2006-01-16

DOI
10.1029/2005gl024975

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Peer reviewed
Late-spring increase of trans-Pacific pollution transport in the upper troposphere

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Received 17 October 2005; revised 10 November 2005; accepted 29 November 2005; published 7 January 2006.

The observations during the Tropospheric Ozone Production about the Spring Equinox (TOPSE) experiment show large enhancements of NOx, PAN, O3, CO, CFCs, and Halon-1211 in the upper troposphere over North America in late spring. Analysis of these observations and model results indicate that the enhancements are most likely driven by a surge of trans-Pacific pollutant transport in late spring. The rapid seasonal transition is particularly striking for upper tropospheric NOx, resulting in large increases in photochemical oxidation and O3 production during the period. The transition is later in season than that of low-altitude trans-Pacific transport, which peaks in March and April. The current generation of global chemical transport models clearly underestimates this long-range transport of pollutants, implying an underestimation in the model-projected impact on regional air quality over North America (through subsidence). Citation: Wang, Y., Y. Choi, T. Zeng, B. Ridley, N. Blake, D. Blake, and F. Flocke (2006), Late-spring increase of trans-Pacific pollution transport in the upper troposphere, Geophys. Res. Lett., 33, L01811, doi:10.1029/2005GL024975.

1. Introduction

Trans-Pacific transport of pollutants to North America has long been recognized [e.g., Andrae et al., 1988; Merrill et al., 1989; Kriz et al., 1990; Parrish et al., 1992]. A particular concern is how increasing pollution from Asia driven by rapid regional economic growth affects O3 concentrations in the United States [e.g., Bernsten et al., 1999; Jacob et al., 1999]. For example, the subsidence of trans-Pacific transported high O3, produced catalytically by NOx (NO + NO2) during the oxidation of CO and volatile organic compounds (VOCs), could significantly contribute to exceedances of the National Ambient Air Quality Standard at California mountain sites [Hudman et al., 2004].

Measurements aimed at exploring the effects of trans-Pacific transport have focused previously on low-altitude O3, CO, and peroxyacetyl nitrate (PAN) [Jaffé et al., 1999; Lin et al., 2000; Jaffé et al., 2003]. In this work, we focus on two aspects that have not been explored in the previous analyses of trans-Pacific transport and evaluate our current capability to simulate these observed features. The first aspect is whether the characteristics of upper tropospheric trans-Pacific transport differ from those at lower altitudes. The second is whether we can deduce the effects of trans-Pacific transport on O3 and the most critical precursor for its production, NOx, over North America. To our knowledge, no previous attempt has been made to examine the impact of trans-Pacific transport on North American NOx.

We focus our analysis on the measurements made during the Tropospheric Ozone Production about the Spring Equinox (TOPSE) experiment during February–May, 2000 [Atlas et al., 2003]. Thirty-eight science flights were conducted in 7 deployments (1–2 weeks apart), covering the region from Colorado to north of Thule, Greenland. A comprehensive suite of chemical species related to tropospheric O3 chemistry was measured from the surface up to 8 km.

To analyze these measurements, we make use of a 3-D regional chemical transport model (RCTM) [Choi et al., 2005] and the global GEOS-CHEM model [Bey et al., 2001]. The RCTM model domain covers the continental United States and Canada with a horizontal resolution of 70 × 70 km2 and 21 layers up to 100 hpa in the vertical [Choi et al., 2005]. The National Center for Atmospheric Research/Penn State MM5 was used to simulate the meteorological fields using four dimensional data assimilation with the National Center for Environmental Prediction reanalysis, surface, and rawinsonde observations. The spring 2000 simulations using the global GEOS-CHEM model (version 7.2.4 with a horizontal resolution of 2° × 2.5° and 30 layers up to 0.01 hpa, GEOS-3 meteorological fields) provide the initial and hourly boundary conditions for trace gases. The RCTM shares the chemistry and deposition modules of GEOS-CHEM. Particular attention was paid to the implementation of convective transport and lightning NOx production schemes in the RCTM [Choi et al., 2005].

2. Results and Discussion

During TOPSE, the largest increases in upper tropospheric reactive nitrogen NOx and PAN were observed during late spring [Wang et al., 2003a]. We first investigate the potential impact of trans-Pacific transport on NOx concentrations. Figure 1 shows the observed and simulated NOx mixing ratios from March to May during TOPSE. The aircraft observations clearly show large increases of NOx concentrations above 5 km from March to May. The RCTM is in reasonable agreement with the observed low concentrations in the upper troposphere in February (not shown).
and March. The model simulates some enhancements in April but none in May, and hence greatly underestimates NOx concentrations. The April enhancements in the RCTM are due to localized convective transport and lightning NOx production. Those enhancements were not simulated in the global GEOS-CHEM model.

[7] Both models show a lack of significant lightning and convective activity over the region in May due to the presence of a high-pressure ridge system residing over the western United States. Inspection of measurements in each flight during May (not shown) finds 4 occasions with NOx mixing ratios > 50 pptv at altitude > 6 km. These enhancements are up to 200 km horizontally and 2 km vertically. The measured dimensions are often limited by sampling. The corresponding model simulations show discernable but much underestimated enhancements for two occasions (not shown). Previous NOx simulations using different regional (HANK [Hess et al., 2000]) and global (MOZART-2 [Horowitz et al., 2003]) CTMs have also shown model underestimations of 50–60% with few data points of simulated NOx mixing ratios > 50 pptv above 5 km [Emmons et al., 2003]. The previous researchers did not speculate on the causes for the large model underestimation.

[8] Overall, four different models with largely independent model formulations of convective transport and lightning parameterization and often drastically different meteorological fields and chemical formulations show that the models greatly underestimate upper tropospheric NOx concentrations in April and May. The cause does not appear to be a poor representation of convection or lightning NO production. Model simulated NOx enhancements due to local convection and associated lightning NOx production are low due to the prevailing high-pressure ridge system over the western United States. In addition, previous comparisons of RCTM simulations with satellite NOx and CO observations indicate that the model captures reasonably well the observed day-to-day variations in lightning NO emissions and convective activity [Choi et al., 2005]. We therefore hypothesize that trans-Pacific transport of pollutants in the upper troposphere into the region was underestimated in the models. The hypothesis can be better tested by examining the observations and simulations of other chemical tracers. We focus on the comparison in May when the pollutant enhancements are most significant.

[9] We compare in Figure 2 the observed and simulated CO, PAN, and O3 concentrations in May. The RCTM has distributions similar to GEOS-CHEM but with more localized enhancements. Hence only the former is shown. Carbon monoxide is a good tracer for anthropogenic emissions. Peroxyacetyl nitrate is the reaction product of NO2 and peroxyacetyl radicals formed during the oxidation of VOCs. It is therefore a good tracer for tropospheric chemical activity of O3 precursors. While in reasonable agreement below 6 km, the simulated CO, PAN, and O3 concentrations are much lower than the observations at higher altitudes. The observed upper tropospheric enhancements of CO, PAN, and O3 tend to be collocated with those of NOx (Figure 1). A close inspection of the simulated O3 concentrations shows slight enhancements in the upper tropospheric regions where high O3 was observed. The enhancements correspond to those in the GEOS-CHEM simulated western boundary conditions and are not caused by photochemical production in the regional model, suggesting that the global model does have some skill in simulating trans-Pacific transport [e.g., Hudman et al., 2004] but the simulated magnitudes are too small.

[10] The discrepancies between simulated and observed CO and PAN concentrations are much larger in May than the three previous months (not shown). At lower altitudes (<6 km), the model reproduces well the observed rapid decrease of CO from April to May. The decrease reflects more active photochemical oxidation toward summer. While the observations show a large increase of CO at higher altitudes (>6 km), the simulated concentrations are less in May than April. The simulated PAN concentrations in May are similar to April at high altitudes, while the observations also show a large increase. The observed O3 mixing ratios at high altitudes show a clear increase from

![Figure 1](image1.png)

**Figure 1.** Observed and RCTM simulated monthly mean distributions of NOx from March to May, 2000. The RCTM data were sampled along the TOPSE flight tracks.

![Figure 2](image2.png)

**Figure 2.** Same as Figure 1 but for CO, PAN, and O3 in May. When constructing the observed O3 distribution, we did not include measurements with mixing ratios >110 ppbv to filter out the effect of stratospheric O3.
of upper tropospheric NOx, CO, PAN, and O3 are due to CO and NOx concentrations through transport, but relatively high O3 concentrations mainly reflect NOx concentrations with high CFC-12. However, the point-to-point correspondence is poor possibly for two reasons. First, CFC-12 was sampled at a much lower frequency than NOx. Secondly, the surface sources of NOx and CFCs are not necessarily collocated and NOx concentrations are also affected by lightning production.

A major limitation of this work is that we do not know the exact origins for the observed trace gas enhancements. While TOPSE measurements are useful to examine the impact of trans-Pacific transport, the effects of specific distant sources become difficult to diagnose. Previous studies indicate that emissions of NOx and CO are too low in China by ~50% [Heald et al., 2004; Wang et al., 2004]. In a sensitivity study (not shown), we doubled the Chinese surface emissions of these two gases. The large underestimation of upper tropospheric NOx in TOPSE regions persists, likely reflecting the low export efficiency from the boundary layer and short chemical lifetime of NOx. Observed upper tropospheric CO enhancements are simulated well north of 50°N but still underestimated at lower latitudes. However, GEOS-CHEM now overestimates CO in February and March. TOPSE measurements do not show a strong increase of CH3Cl in May implying that the contribution from biomass burning to the seasonal increase is limited. It is possible that trans-Pacific pollutant transport surges in May due to a significant increase of convection and lightning over East Asia or the western Pacific (not simulated in the models). Detailed analysis of other measurements will be necessary to explore that possibility.

3. Conclusions

TOPSE observations show large enhancements of NOx, PAN, CO, O3, and CFCs at altitude > 6 km in May. We hypothesize that these enhancements are due to trans-Pacific transport. The hypothesis is supported by our analysis of the observations and model results. First, we find that these chemical tracers show consistent enhance-
ment patterns at high altitudes. Both CFC enhancements and backtrajectory calculations imply trans-Pacific transport from East Asia. Secondly, the observed NO$_2$-O$_3$ correlation for high CO data points indicates significant tropospheric photochemical production. Thirdly, the relatively high PAN/NO$_2$ ratios of >5 indicate photochemically aged air masses. Lastly, separate regional/global chemical transport models using different meteorological fields and chemical formulations consistently underestimate the enhancements of NO$_x$, PAN, and CO. The contribution by North American surface emissions is limited in these simulations because the high-pressure ridge system over the western United States suppresses convection and lightning in May.

[16] Our results indicate that the rapid late-spring increase of reactive nitrogen NO$_x$ and PAN at northern mid-latitude upper troposphere observed during TOPSE is most likely due to the enhancements of these species by trans-Pacific transport. These enhancements result in significant increases of photochemical oxidation and O$_3$ production. The trace gas enhancements including that of CO are still increasing in May. It is very different from trans-Pacific transport driven CO enhancements at low altitudes, which peak in March and April [e.g., *Weiss-Penzias et al.*, 2004]. The current global CTMs do not capture this rapid seasonal transition, which results in a large underestimation of photochemical production of O$_3$ in the models. The problem raises concerns on our capability to assess the effects of intercontinental transport on regional air quality.

[17] Acknowledgments. This work was supported by the National Science Foundation Atmospheric Chemistry Program. The GEOS-CHEM model is managed at Harvard University with support from the NASA Atmospheric Chemistry Modeling and Analysis Program; we thank Robert Yantosca and Daniel Jacob for providing us the model and data. NCAR is operated by the University Corporation for Atmospheric Research under sponsorship of the National Science Foundation.

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