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Authors

Heald, Colette L
Jacob, Daniel J
Fiore, Arlene M
[et al.](#)

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Asian outflow and trans-Pacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft, and model perspective

Colette L. Heald,¹ Daniel J. Jacob,¹ Arlene M. Fiore,¹ Louisa K. Emmons,² John C. Gille,² Merritt N. Deeter,² Juying Warner,² David P. Edwards,² James H. Crawford,³ Amy J. Hamlin,^{3,4} Glen W. Sachse,³ Edward V. Browell,³ Melody A. Avery,³ Stephanie A. Vay,³ David J. Westberg,⁵ Donald R. Blake,⁶ Hanwant B. Singh,⁷ Scott T. Sandholm,⁸ Robert W. Talbot,⁹ and Henry E. Fuelberg¹⁰

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[1] Satellite observations of carbon monoxide (CO) from the Measurements of Pollution in the Troposphere (MOPITT) instrument are combined with measurements from the Transport and Chemical Evolution Over the Pacific (TRACE-P) aircraft mission over the northwest Pacific and with a global three-dimensional chemical transport model (GEOS-CHEM) to quantify Asian pollution outflow and its trans-Pacific transport during spring 2001. Global CO column distributions in MOPITT and GEOS-CHEM are highly correlated ($R^2 = 0.87$), with no significant model bias. The largest regional bias is over Southeast Asia, where the model is 18% too high. A 60% decrease of regional biomass burning emissions in the model (to 39 Tg yr⁻¹) would correct the discrepancy; this result is consistent with TRACE-P observations. MOPITT and TRACE-P also give consistent constraints on the Chinese source of CO from fuel combustion (181 Tg CO yr⁻¹). Four major events of trans-Pacific transport of Asian pollution in spring 2001 were seen by MOPITT, in situ platforms, and GEOS-CHEM. One of them was sampled by TRACE-P (26–27 February) as a succession of pollution layers over the northeast Pacific. These layers all originated from one single event of Asian outflow that split into northern and southern plumes over the central Pacific. The northern plume (sampled at 6–8 km off California) had no ozone enhancement. The southern subsiding plume (sampled at 2–4 km west of Hawaii) contained a 8–17 ppbv ozone enhancement, driven by decomposition of peroxyacetyl nitrate (PAN) to nitrogen oxides (NO_x). This result suggests that PAN decomposition in trans-Pacific pollution plumes subsiding over the United States could lead to significant enhancements of surface ozone. *INDEX TERMS*: 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*: TRACE-P, MOPITT, Asian pollution, trans-Pacific transport, carbon monoxide, O₃ production

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1. Introduction

[2] Quantifying the continental outflow and intercontinental transport of air pollutants is one of the major

challenges of atmospheric chemistry today. Integration of satellite and aircraft observations with three-dimensional (3-D) models is a promising new approach for addressing this challenge. We present here such an integration through a combined model interpretation of aircraft observations of

¹Department of Earth and Planetary Sciences, Harvard University, Cambridge, Massachusetts, USA.

²National Center for Atmospheric Research, Boulder, Colorado, USA.

³NASA Langley Research Center, Hampton, Virginia, USA.

⁴Now at College of Engineering, Michigan Technological University, Houghton, Michigan, USA.

⁵Science Applications International Corporation, Hampton, Virginia, USA.

⁶Department of Chemistry, University of California, Irvine, California, USA.

⁷NASA Ames Research Center, Moffet Field, California, USA.

⁸School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, Georgia, USA.

⁹Institute for the Study of Earth, Oceans and Space, University of New Hampshire, Durham, New Hampshire, USA.

¹⁰Department of Meteorology, Florida State University, Tallahassee, Florida, USA.

Asian pollution outflow and trans-Pacific transport from the Transport and Chemical Evolution Over the Pacific (TRACE-P) aircraft mission, together with satellite observations of carbon monoxide (CO) from the Measurement of Pollution in the Troposphere (MOPITT) instrument. In the process we demonstrate the value of the MOPITT observations for mapping intercontinental transport of pollution, and we present the first observationally based analysis of the production of ozone during trans-Pacific transport of Asian pollution.

[3] The NASA TRACE-P mission focused on the study of Asian chemical outflow over the northwest Pacific in February–April 2001 [Jacob *et al.*, 2003]. Two aircraft intensively sampled the region from bases in Hong Kong and Japan. Transit flights from the United States sampled a major trans-Pacific pollution transport event. The MOPITT instrument [Drummond and Mand, 1996; Edwards *et al.*, 1999] provided global coverage of CO concentrations from a polar orbit. Validation profiles for MOPITT conducted by the TRACE-P aircraft [Jacob *et al.*, 2003] set the stage for a combined analysis of the TRACE-P and MOPITT data sets. A global chemical transport model is used here to provide the necessary transfer function between the MOPITT and aircraft observations, and to place these observations in the context of our current understanding of sources and transport from Asia.

[4] Carbon monoxide, a product of incomplete combustion, is present at concentrations of 50–500 ppbv in the troposphere. It is the principal sink of OH, the main tropospheric oxidant, and as such plays a key role in determining the oxidizing power of the atmosphere. It has a mean atmospheric lifetime of 2 months against oxidation by OH, its main atmospheric sink, and is therefore also a sensitive tracer for continental outflow and intercontinental transport of pollution [Staudt *et al.*, 2001; Liu *et al.*, 2003].

[5] Recent studies have suggested that trans-Pacific transport of Asian pollution has significant implications for ozone and aerosol air quality in the United States [Berntsen *et al.*, 1999; Jaffe *et al.*, 1999; Jacob *et al.*, 1999; Yienger *et al.*, 2000; Fiore *et al.*, 2002]. This influence could be magnified during synoptic events such as those documented during the Photochemical Ozone Budget of the Eastern North Pacific Atmosphere (PHOEBEA) aircraft campaigns [Jaffe *et al.*, 1999, 2001; Jaeglé *et al.*, 2003; Price *et al.*, 2003; Jaffe *et al.*, 2003]. Yienger *et al.* [2000] have suggested that three to five large Asian pollution events reach the western United States every spring. Asian pollution is typically exported to the Pacific via frontal lifting in warm conveyor belts (WCBs) or convection to the free troposphere [Bey *et al.*, 2001b; Husar *et al.*, 2001; Stohl, 2001; Liu *et al.*, 2003] and can then be transported across the Pacific in 5–10 days [Jaffe *et al.*, 1999; Yienger *et al.*, 2000; Jaffe *et al.*, 2001; Stohl *et al.*, 2002]. The evolution and characteristics of one such event will be examined here, in the larger context of the trans-Pacific pollution transport observed by MOPITT throughout the TRACE-P mission.

2. Observations and Model

2.1. TRACE-P Observations

[6] The TRACE-P mission involved two NASA aircraft (DC-8 and P3-B) deployed from bases in Hong Kong and

Japan to sample outflow from Asia. Transit flights from the United States sampled a major event of trans-Pacific transport of Asian pollution on 26 and 27 February, described in section 4. The aircraft payload included instrumentation to measure O₃, CO, reactive nitrogen oxide (NO_y) species, and a suite of hydrocarbon and halocarbon tracers [Jacob *et al.*, 2003]. Observations of CO were made continuously with 1 Hz frequency using the Differential Absorption of CO Measurement (DACOM) instrument [Sachse *et al.*, 1987]. The measurement accuracy is approximately 2%.

[7] Fuelberg *et al.* [2003] give an overview of the meteorological conditions during TRACE-P. Asian pollution was primarily exported to the Pacific in WCBs ahead of cold fronts [Liu *et al.*, 2003]. This pollution included both anthropogenic (defined herein as fossil fuel and biofuel sources only) and biomass burning influences, the latter due to seasonal biomass burning in Southeast Asia [Heald *et al.*, 2003]. The two influences were often mixed in the outflow [Carmichael *et al.*, 2003; Ma *et al.*, 2003]. Following standard practice we distinguish “anthropogenic” from “biomass burning” sources, even though biomass burning here is largely due to human activity. The distinction is useful because biomass burning has a separate geographical and seasonal signature, and because the inventories are constructed from very different types of bottom-up information.

[8] Detailed inventories for anthropogenic emissions [Streets *et al.*, 2003] and daily biomass burning emissions [Heald *et al.*, 2003] were constructed for the TRACE-P period from bottom-up economic and fire information, respectively. These inventories were used by Palmer *et al.* [2003] in an inverse model analysis to determine the constraints from the TRACE-P CO observations toward better quantifying CO sources from east Asia. They concluded that anthropogenic emissions from China in the Streets *et al.* [2003] inventory were underestimated by 35%, and that biomass burning emissions from Southeast Asia in the Heald *et al.* [2003] inventory were a factor of three too high.

[9] Other TRACE-P studies examined the Asian outflow of ozone and of NO_y. Large NO_y enhancements were observed downwind from Asia and consisted mainly of HNO₃ and PAN [Miyazaki *et al.*, 2003; Talbot *et al.*, 2003]. PAN was the dominant form of NO_y in outflow associated with WCBs [Miyazaki *et al.*, 2003]. Examination of NO_y-CO correlations by Koike *et al.* [2003] indicated an NO_y export efficiency to the free troposphere from northeastern China of 15%, with only 0.5% of the emitted NO_x remaining as NO_x in the free tropospheric outflow. Significant ozone production in Asian outflow during TRACE-P appeared to be limited to biomass burning plumes originating from low latitudes [Tang *et al.*, 2003]. Ozone production in anthropogenic plumes was generally insignificant, reflecting the weak photochemical activity at that time of year [Pierce *et al.*, 2003].

2.2. MOPITT Observations

[10] The MOPITT instrument is a nadir IR correlation radiometer launched aboard the NASA Earth Observing System Terra spacecraft in 1999 [Drummond, 1992; Edwards *et al.*, 1999]. It has solar backscatter channels to measure total CO columns, and IR emission channels to measure CO vertical profiles. Correlation spectroscopy

provides two signals for each channel. So far only the IR emission channels have been retrieved. The satellite is deployed in a polar Sun-synchronous orbit with a 10:30 local equator crossover time. Horizontal resolution is $22 \text{ km} \times 22 \text{ km}$, and cross-track scanning achieves approximate global coverage in 3 days. CO concentrations are reported on 7 vertical levels (surface, 850, 700, 500, 350, 250, and 150 hPa), and as a total column, for all cloud-free scenes.

[11] The MOPITT observations reported here are version 3 data (<http://www.eos.ucar.edu/mopitt>), which have a 10% precision on both column and mixing ratio [Pan *et al.*, 1998; Edwards *et al.*, 2003]. Current CO retrievals are based on only 4 signals which limits the vertical resolution, as described below [Deeter *et al.*, 2003]. The retrieval is ill conditioned without the inclusion of external information in the form of an a priori profile \mathbf{x}_a and its error covariance matrix \mathbf{S}_a . The vertical profile $\hat{\mathbf{x}}$ retrieved by the instrument is related to the true vertical profile \mathbf{x} by the averaging kernel matrix \mathbf{A} , as described by Rodgers [2000]:

$$\hat{\mathbf{x}} = \mathbf{x}_a + \mathbf{A}(\mathbf{x} - \mathbf{x}_a) + \mathbf{G}\boldsymbol{\epsilon}, \quad (1)$$

where $\boldsymbol{\epsilon}$ is the measurement error and \mathbf{G} is the gain matrix. The MOPITT retrieval uses a single profile of \mathbf{x}_a (and \mathbf{S}_a) with mixing ratio decreasing from 120 ppbv at the surface to 75 ppbv at 250 hPa and less than 60 ppbv at 150 hPa [Deeter *et al.*, 2003]. The averaging kernel matrix is given by

$$\mathbf{A} = \mathbf{I} - \hat{\mathbf{S}}\mathbf{S}_a^{-1}, \quad (2)$$

where \mathbf{I} is the identity matrix and $\hat{\mathbf{S}}$ is the retrieval error covariance matrix, different for each retrieval and supplied with the MOPITT retrieval product. The rows of \mathbf{A} (averaging kernels) describe the vertical resolution of the retrieval. A sample set of averaging kernels for the North Pacific is shown in Figure 1. The column averaging kernel (right panel of Figure 1) specifies the total column sensitivity to a change in the true mixing ratio at each level and is calculated as follows:

$$\mathbf{a} = \mathbf{t}^T \mathbf{A}, \quad (3)$$

where \mathbf{t} is the transfer operator which includes both the conversion from volume mixing ratio to number density and the integration over pressure. Figure 1 shows roughly two independent regions of information in the MOPITT retrieved profile: one in the middle troposphere (300–800 hPa) and one in the upper troposphere (pressure < 500 hPa) [Deeter *et al.*, 2003]. For details on how the averaging kernels vary with land types and time of day, see Deeter *et al.* [2003]. We find that the global average number of degrees of freedom for signal in the MOPITT vertical profiles, which is the total of the eigenvalues of the averaging kernel and describes the number of independent pieces of information inferred from the retrieval [Rodgers, 2000], is 1.21 ± 0.45 for 1 sample day (15 March 2001). One can think of this value as the number of modes of vertical structure retrieved by MOPITT [Rodgers, 1990]. The first eigenmode for MOPITT is representative of a broad column-like signal with an eigenvalue close to one.

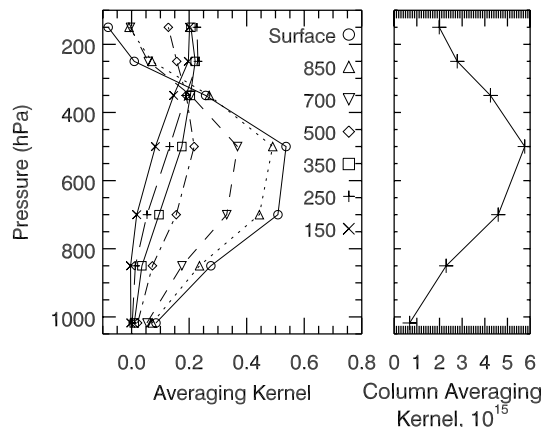


Figure 1. Measurements of Pollution in the Troposphere instrument averaging kernels for a sample retrieval over the North Pacific (28.2°N , 139.3°E , 23 February 2001). Each curve in the left panel represents a row of the averaging kernel matrix and describes the sensitivity of retrieved CO at a particular pressure level (indicated in the legend of the figure in units of hPa) to perturbations to the a priori profile at different pressures (ordinate on the figure). The column averaging kernel is shown in the right panel.

The remaining contribution to d_s , which can exceed 0.5 in the tropics but is generally less than 0.2 in the extratropics, can be thought of as the contribution from a broad vertical gradient. We choose to focus on the column data in our analysis.

[12] Seven MOPITT validation profiles for a range of conditions were performed during TRACE-P underneath coincident MOPITT overpasses. Four were conducted under sufficiently clear-sky conditions to enable MOPITT retrievals. Results presented by Jacob *et al.* [2003] show that MOPITT captures the variability in the observed aircraft columns, with a high bias of $6 \pm 2\%$, but provides no apparent information on vertical structure for Asian plumes over the Pacific [Jacob *et al.*, 2003]. One of the validation profiles (DC-8 flight 5) sampled a trans-Pacific pollution transport layer event with CO in excess of 250 ppbv, as discussed in section 3. The 6% MOPITT bias observed during TRACE-P is consistent with MOPITT validation results for a suite of aircraft campaigns and profiles (L. Emmons *et al.*, Validation of MOPITT CO retrievals with aircraft in situ profiles, submitted to *Journal of Geophysical Research*, 2003).

2.3. Model Description

[13] We use the GEOS-CHEM v. 4.33 global 3-D model of tropospheric chemistry (<http://www-as.harvard.edu/chemistry/trop/geos>) with $2^\circ \times 2.5^\circ$ horizontal resolution and 48 vertical levels. The model is driven by assimilated meteorology from the GEOS-3 of the NASA Data Assimilation Office and was initially described by Bey *et al.* [2001a]. The GEOS-3 data have a temporal resolution of 6 hours (3 hours for surface variables and mixing depths). Other TRACE-P studies using the GEOS-CHEM model have demonstrated the ability of the model to simulate the meteorological and chemical signatures of Asian outflow over the Pacific [Jaeglé *et al.*, 2003; Kiley

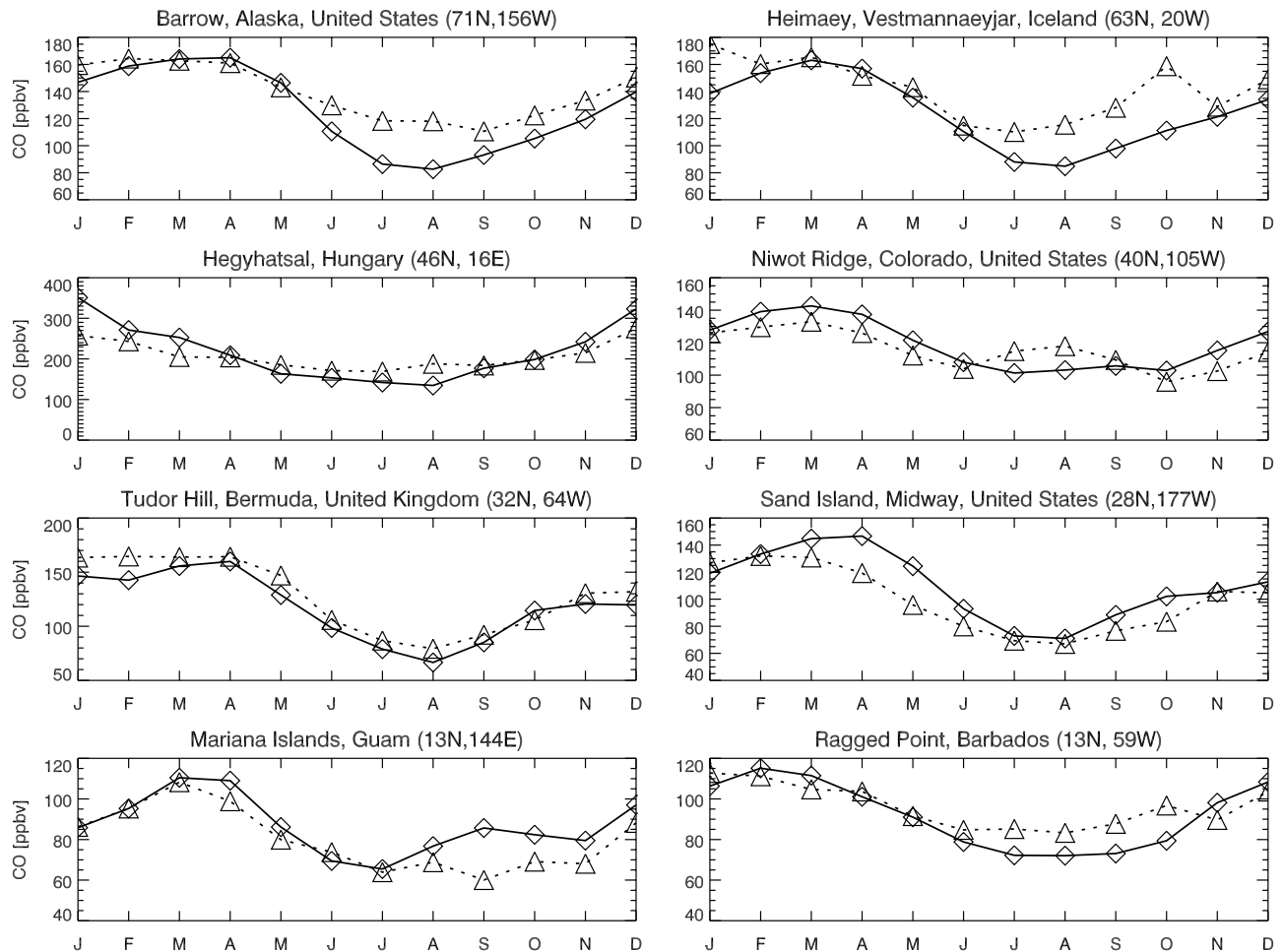


Figure 2. Comparisons of observed (solid lines) and simulated (dotted lines) monthly mean CO concentrations for 2001 at selected Northern Hemispheric Climate Monitoring and Diagnostics Laboratory surface stations.

et al., 2003; *Li et al.*, 2003; *Liu et al.*, 2003; *Palmer et al.*, 2003].

[14] We conducted a simulation for February–April 2001 with full O_3 - NO_x -hydrocarbon chemistry. We also conducted CO-only simulations with tagged sources and monthly mean OH concentration fields archived from the full chemistry simulation. Initial conditions were taken from a 1-year spin-up simulation and model output was sampled every 3 hours. Anthropogenic sources of CO and its precursor hydrocarbons are as described by B. N. Duncan *et al.* (Model study of the variability and trends of carbon monoxide (1988–1997): 1. Model formulation, evaluation, and sensitivity, submitted to *Journal of Geophysical Research*, 2003, hereinafter referred to as Duncan *et al.*, submitted manuscript, 2003). Our anthropogenic source of CO from China ($181 \text{ Tg CO yr}^{-1}$) is 66% higher than in the inventory of *Streets et al.* [2003] and 8% higher than the a posteriori estimate from the inverse model by *Palmer et al.* [2003] constrained with TRACE-P observations. Biomass burning emissions are from a bottom-up inventory constrained with daily satellite fire counts for the TRACE-P period [*Heald et al.*, 2003]. Most of the global biomass burning for this period takes place in Southeast Asia and the corresponding

regional source for February–April 2001 is 69 Tg CO . The inverse model analysis of *Palmer et al.* [2003] suggests that this regional biomass burning source is too high by a factor of 3; as we will see, simulation of the MOPITT data leads to a similar conclusion.

[15] Comparisons between full-year model results for 2001 and ground station CO measurements by the National Ocean and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL) Carbon Cycle Group for representative Northern Hemispheric sites are shown in Figure 2. Several recent changes to the GEOS-CHEM chemical mechanism, including a faster $N_2 + O(^1D)$ rate constant [*Ravishankara et al.*, 2002] have decreased the global mean OH concentration by 10–15%. The most recent global model evaluation of CO by Duncan *et al.* (submitted manuscript, 2003) which does not include this change in rate constant, shows a model underestimate in the northern tropics and midlatitudes by 10–40 ppbv, with better agreement achieved in the arctic latitudes. The model simulation reported here is generally unbiased in its simulation of the CMDL Northern Hemispheric sites; however high-latitude sites are overestimated by 10–30 ppbv in summer-fall. Our 10–30 ppbv spring underestimate at Midway (MID) does not appear to be driven by an underestimate in Asian sources

since neither the TRACE-P observations nor the MOPITT observations (discussed below) reveal such a bias.

3. Interpretation of MOPITT Observations for the TRACE-P Period

3.1. Global Distributions

[16] Figure 3 shows the mean CO column observed by MOPITT during TRACE-P (20 February to 10 April 2001) and the corresponding GEOS-CHEM model column with MOPITT averaging kernels applied (according to equation (1)). Here the MOPITT data for each day have been averaged over the $2^\circ \times 2.5^\circ$ GEOS-CHEM model grid and the resulting gridded product is then averaged over the 49-day TRACE-P period. MOPITT retrievals are not performed for cloudy scenes and are limited by the orbital coverage, but the effective coverage is increased here because of averaging over the $2^\circ \times 2.5^\circ$ grid. Temporal coverage of the gridded MOPITT product over the North Pacific is 20–70% (Figure 4). The GEOS-CHEM model results in Figure 3 were constructed by binning the model on the MOPITT pressure levels for each retrieval scene, weighting by the MOPITT averaging kernels (such as shown in Figure 1), and integrating the column.

[17] Both model and satellite observations show high CO levels over continental source and outflow regions, as well as a general latitudinal gradient. Columns are low over high terrain (e.g., the Himalayas, the Rocky Mountains). A difference plot (Figure 3, bottom panel) shows no mean global bias between the model and measured CO. Mean MOPITT and GEOS-CHEM columns for the TRACE-P period are highly correlated ($R^2 = 0.97$, slope = 0.97); and the correlation also holds for data points on individual days ($R^2 = 0.87$, slope = 0.95; Figure 5). The slope indicates that the model underestimates MOPITT observations by 5%. As pointed out in section 2.2, the MOPITT observations appear to be biased high by 6% relative to the TRACE-P aircraft data.

3.2. Constraints on Asian Emissions

[18] The pattern of model bias relative to MOPITT matches the distribution of biomass burning, notably over Southeast Asia [Heald *et al.*, 2003]. MOPITT column observations over Southeast Asia (10° – 25° N, 80° – 110° E), where biomass burning dominates anthropogenic emissions by a factor of two in the model, are on average 18% lower than the model column. The bias extends to southern China, where biomass burning effluents from Southeast Asia are lifted to the middle troposphere during transport in WCBs [Liu *et al.*, 2003]. The discrepancy suggests that biomass burning emissions in the model for Southeast Asia are too high, consistent with the inverse model analysis of the TRACE-P CO data by Palmer *et al.* [2003]. In order to estimate how the overestimate of MOPITT observations translates to an overestimate of biomass burning emissions, we reduced the mean simulated CO profile over Southeast Asia by a fraction of the mean biomass burning contribution (determined from a sensitivity simulation). We then passed the resulting profile through the averaging kernels for 1 sample day (16 March 2001). We thus estimate in the model that reducing the biomass burning source over Southeast Asia by 60% (to 39 Tg yr^{-1} CO over the 70° – 140° E, 0° – 50° N region) would

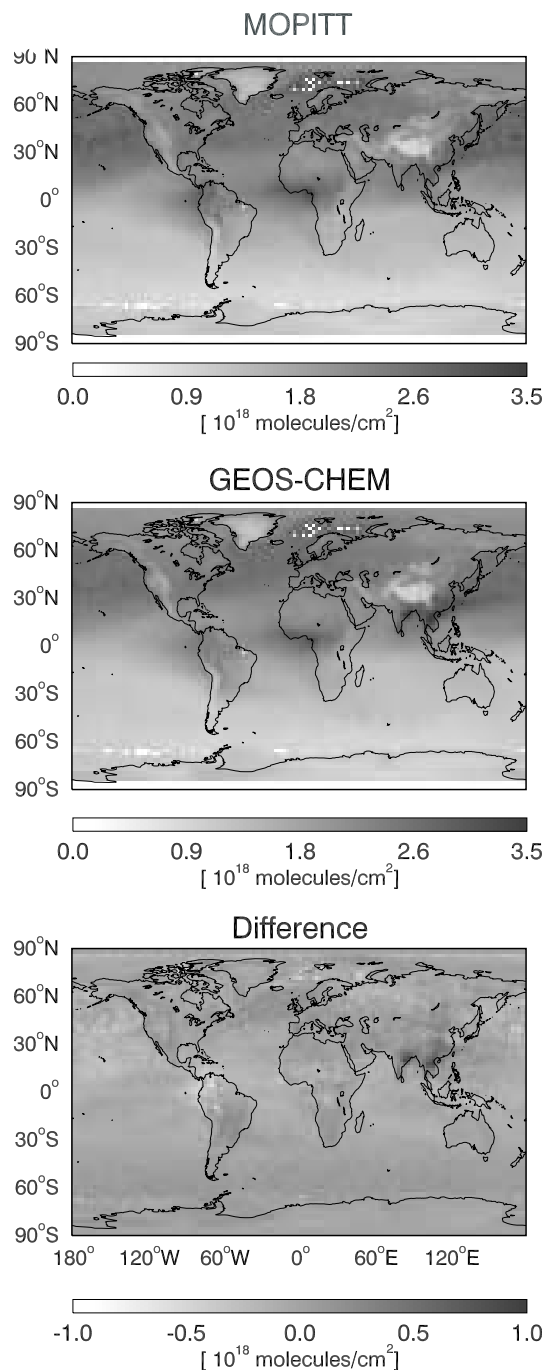


Figure 3. Mean CO columns measured by MOPITT during the Transport and Chemical Evolution Over the Pacific (TRACE-P) campaign (20 February to 10 April 2001) and corresponding GEOS-CHEM model columns sampled along the MOPITT observational track and transformed with the MOPITT averaging kernels. Both the MOPITT data and the transformed GEOS-CHEM fields have been averaged over the GEOS-CHEM $2^\circ \times 2.5^\circ$ model grid. The bottom panel shows the difference between GEOS-CHEM and MOPITT. The color scales for the top two panels are saturated at 3.5×10^{18} molecules/cm², and the bottom panel scale is saturated at 1.0×10^{18} molecules/cm². See color version of this figure at back of this issue.

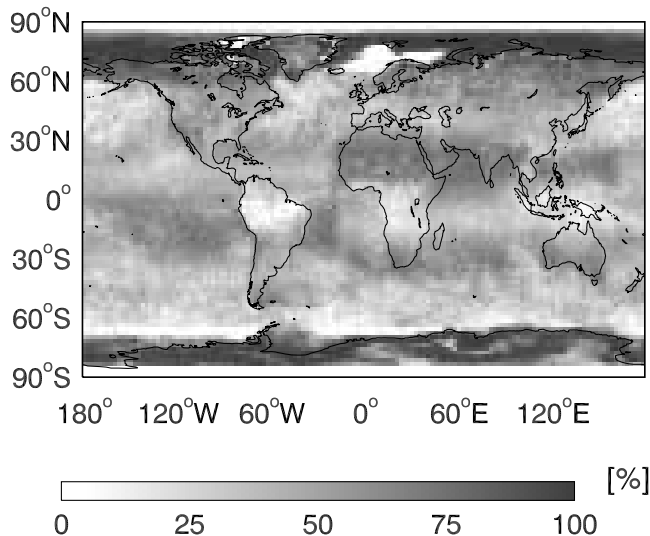


Figure 4. Percentage of days with MOPITT observations in each $2^\circ \times 2.5^\circ$ GEOS-CHEM model grid square during the TRACE-P period (20 February to 10 April 2001). See color version of this figure at back of this issue.

correct the MOPITT overestimate, consistent with the previous inverse analysis of TRACE-P data by *Palmer et al.* [2003]. An important caveat in the interpretation of model vs. MOPITT discrepancies in terms of errors on CO sources is the possibility of model bias in the vertical structure of CO. This could arise, in particular, as a result of

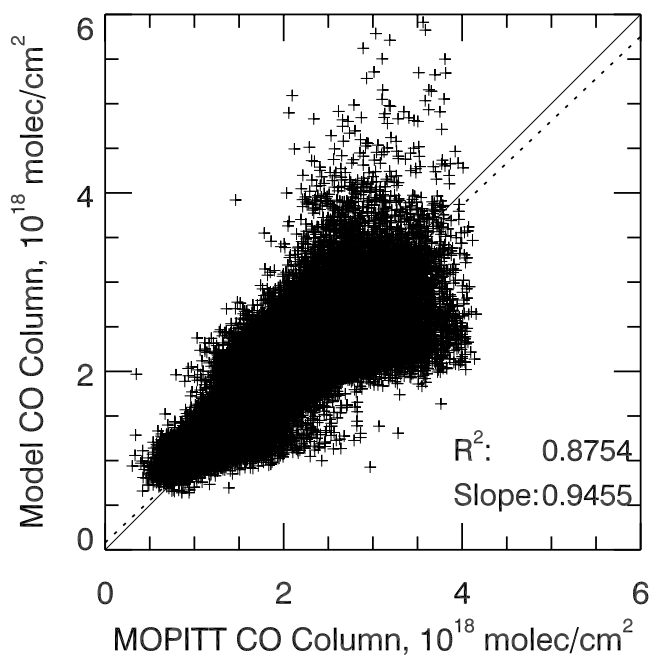


Figure 5. Point-by-point global comparison of daily gridded MOPITT observations of CO columns and corresponding GEOS-CHEM model results for 20 February to 10 April 2001. Also shown is the linear regression (dotted line) and the 1:1 relationship (solid line). The slopes of the linear regression and the corresponding R^2 are indicated. Statistics were computed with the reduced-major-axis method [*Hirsch and Gilroy, 1984*].

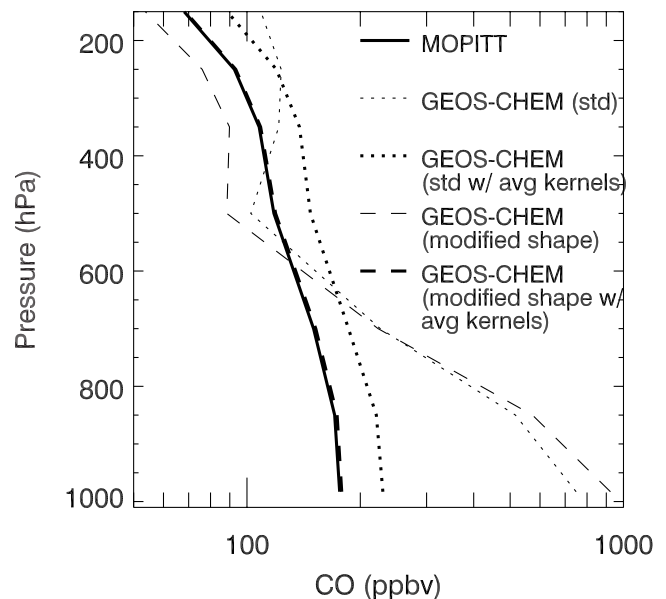


Figure 6. Vertical profiles of CO concentrations over a typical Southeast Asia location (15.6°N , 102.7°E , 23 February 2001) illustrating the sensitivity of the MOPITT retrieval to the vertical distribution of CO. The solid line shows the MOPITT retrieval, the thin dotted line shows results from the standard GEOS-CHEM simulation, and the bold dotted line shows the same GEOS-CHEM results with MOPITT averaging kernels applied. The regional model overestimate is discussed in the text and could reflect excessive biomass burning in the model. However, modifying the shape of the GEOS-CHEM vertical distribution of CO while keeping the CO column constant can also correct the discrepancy. The thin dashed line shows the GEOS-CHEM vertical profile with the modified shape, and the bold dashed line shows the same profile with MOPITT averaging kernels applied.

excessive convection transporting CO to the upper troposphere where MOPITT is most sensitive. Comparison of GEOS-CHEM and MOPITT vertical profiles over Southeast Asia shows no significant difference in shape (Figure 6), but this reflects the lack of MOPITT information on vertical structure as previously discussed. The model vertical profiles over Southeast Asia show an enhancement in the upper troposphere due to deep convection. If we were to change the shape of the vertical profile in the model while conserving the total CO column, such that we reduce upper tropospheric CO to tropical background concentrations (55–70 ppbv) and correspondingly increase lower-tropospheric CO, we would match the MOPITT observations (Figure 6). Cloud data show that frequent deep convection did in fact occur over Southeast Asia during TRACE-P [*Fuelberg et al., 2003*], and an intercomparison of model simulations of CO for the TRACE-P period reveals no systematic bias in the GEOS-CHEM vertical distribution [*Kiley et al., 2003*], but in the absence of measured vertical profiles over Southeast Asia some ambiguity remains.

[19] The MOPITT observations reveal no evident model bias in the anthropogenic source of CO from China. This is consistent with the TRACE-P aircraft observations, since the source used here is only 8% higher than the a posteriori

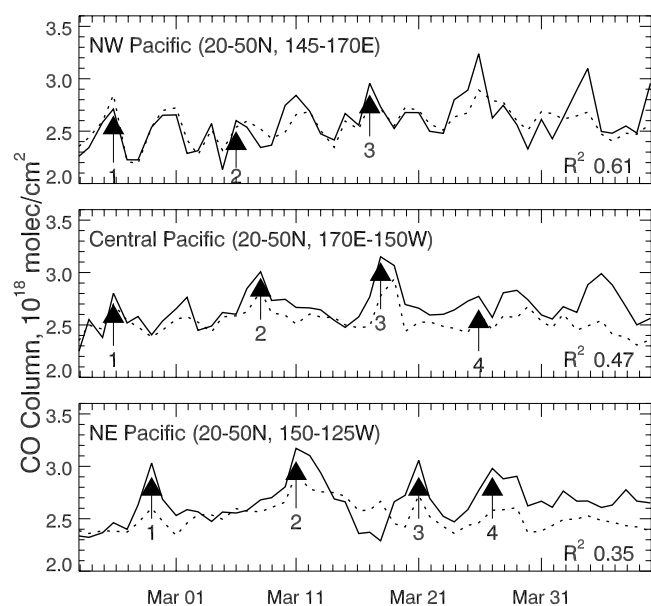


Figure 7. Spring 2001 time series of the average $2^\circ \times 2.5^\circ$ gridded CO column observed by MOPITT (solid line) and simulated by GEOS-CHEM with the averaging kernel applied (dotted line) over the western, central, and eastern North Pacific. Plumes transported across the Pacific are identified and numbered. Event 4 was observed from aircraft during the Photochemical Ozone Budget of the Eastern North Pacific Atmosphere (PHOBEA) II campaign and was characterized as an air mass that had descended from high latitudes but had originated from east Asian sources (Jaffe et al., submitted manuscript, 2002). We were unable to associate this event in the NE Pacific with any specific outflow event in the NW.

estimate derived by Palmer et al. [2003] by inverse modeling of the TRACE-P data. Previous inverse model studies by Kasibhatla et al. [2002] and Pétron et al. [2003] required high anthropogenic emissions from eastern Asia ($350\text{--}380$ Tg yr⁻¹ and >500 Tg yr⁻¹, respectively, compared to our 323 Tg yr⁻¹) to achieve agreement with global CMDL ground station observations in 1994 and 1990–1996, respectively. The constraints from TRACE-P and MOPITT observations imply a weaker Asian source.

3.3. Asian Outflow and Trans-Pacific Transport

[20] We now turn to the variability of Asian outflow to the Pacific and subsequent trans-Pacific transport, as observed by MOPITT and simulated by GEOS-CHEM. Figure 7 shows MOPITT and GEOS-CHEM time series for the TRACE-P period over the northwest, central and northeast Pacific. As before, the GEOS-CHEM model results are sampled for each MOPITT scene and are processed through the MOPITT averaging kernels. The MOPITT observations over the northwest Pacific show outflow events every 5–8 days associated with the passage of cold fronts across the Asian Pacific Rim, as also observed during TRACE-P [Fuelberg et al., 2003; Liu et al., 2003]. GEOS-CHEM and MOPITT are correlated ($R^2 = 0.61$) in this region, reflecting the ability of the model to capture the WCBs driving the Asian outflow ahead of the frontal passages

[Liu et al., 2003]. Farther downwind across the Pacific the agreement between model and observations degrades ($R^2 = 0.35$ over the northeast Pacific), possibly due to accumulation of model transport errors or to vertical displacement of the Asian pollution layers (J. H. Crawford et al., Exploring the relationship between MOPITT and in situ observations of CO based on a large-scale feature sampled during TRACE-P, submitted to *Journal of Geophysical Research*, 2003). Column concentrations are most sensitive to CO enhancements in the midtroposphere (5–6 km) due to the application of the MOPITT averaging kernels (Figure 1). The MOPITT data over the northeast Pacific identify four major events of trans-Pacific transport of Asian pollution during the TRACE-P period, and these are indicated by arrows in Figure 7. These events are all captured by the GEOS-CHEM model but with a dampened magnitude. The four trans-Pacific transport events identified by MOPITT during the TRACE-P period were also seen by in situ observations. Event 1 was observed on the outbound TRACE-P transit flights, as discussed further in section 4. Events 2 and 3 were observed at the Cheeka Peak Observatory (CPO, 48.30°N 124.62°W) on the coast of Washington state on 11 and 20 March, respectively [Jaeglé et al., 2003]. There were no observations at CPO during events 1 and 4. Event 4 was observed from aircraft during the PHOBEA II experiment and was characterized as an air mass that had descended from high latitudes but originated from east Asian sources [Jaffe et al., 2003].

[21] Crawford et al. [2003] showed that high CO concentrations observed over the northwest Pacific during TRACE-P were correlated with cloud cover, reflecting the role of WCBs as the dominant pathway of outflow. They suggested that this might lead to bias in satellite observations of continental outflow. To investigate this issue we compared the average CO column from GEOS-CHEM over the northwest Pacific during the TRACE-P period ($2.62 \pm 0.11 \times 10^{18}$ molecules cm⁻²) to the average of the subset corresponding to MOPITT observation points ($2.57 \pm 0.16 \times 10^{18}$ molecules cm⁻²). There is no significant difference between these two numbers, implying that at least in the model world there is no cloud bias in the MOPITT sampling of the northwest Pacific; however, the model representation of clouds is not particularly robust.

4. Ozone Production During Trans-Pacific Transport: A Case Study

4.1. Trans-Pacific Transport Event on 22–27 February

[22] Layers with elevated CO from rapid trans-Pacific transport of Asian pollution were observed on the two outbound transit flights of the DC-8 during TRACE-P (flight tracks shown in Figure 8). In flight 4 from California to Hawaii (26 February 2001), vertical profiles indicated CO concentrations in excess of 200 ppbv in midtropospheric layers at 600–250 hPa (Figure 9, profiles A, B, and C; locations shown in Figure 8). The southern layers (B and C) were also enhanced in O₃, whereas the northern layer (A) at 40°N off the coast of California was not. In flight 5 from Hawaii to Guam conducted the following day (27 February 2001), strong CO and O₃ enhancement layers were observed in the lower free troposphere at 800–600 hPa (Figure 9, profiles D and E). CO concentrations

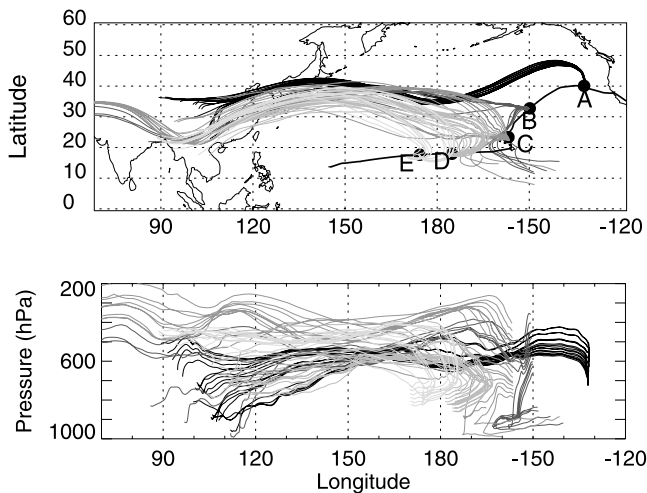


Figure 8. Five-day kinematic back trajectories based on European Centre for Medium-Range Weather Forecasts meteorological fields [Fuelberg *et al.*, 2003] for the enhanced CO layers observed in TRACE-P DC-8 flight 4 from California to Hawaii (26 February) and flight 5 from Hawaii to Guam (27 February). The flight tracks are shown in black, and arrival points in profiles A-E of Figure 9 are shown in red, green, purple, blue, and cyan, respectively. See color version of this figure at back of this issue.

were greater than 200 ppbv, and were accompanied by O_3 in excess of 70 ppbv.

[23] We find that the pollution layers observed in flights 4 and 5 all originated from one single Asian outflow plume on

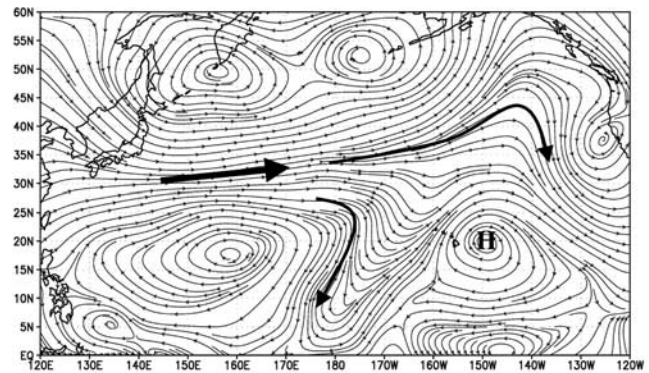


Figure 10. Streamlines at 500 hPa for 25 February, 1800 UTC, indicating the blocking high-pressure system over the north-central Pacific that led to splitting of the Asian pollution plume sampled on TRACE-P DC-8 flights 4 and 5. (NCEP reanalysis data was provided by the NOAA-CIRES Climate Diagnostics Center, Boulder, Colorado, USA, from their Web site at <http://www.cdc.noaa.gov>). The arrows illustrate the transport and splitting of the Asian pollution plume.

22 February 2001 associated with a WCB lifting Chinese pollution ahead of a cold front. Back trajectories (Figure 8) offer evidence for this common origin; further evidence from chemical tracer observations and the GEOS-CHEM model simulation will be presented below. The Asian plume was rapidly transported across the northwest Pacific and encountered a blocking high-pressure system near Hawaii on 25 February (Figure 10). It then split into northern and

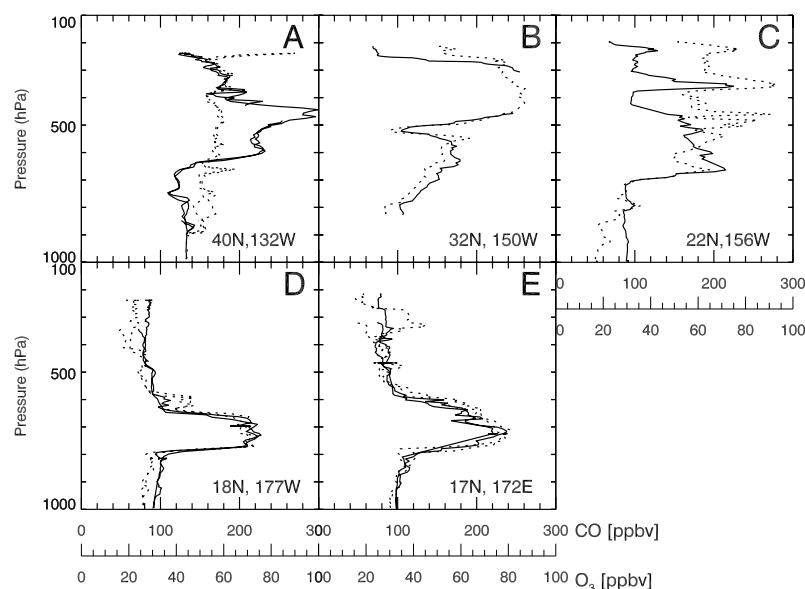


Figure 9. Vertical concentration profiles of CO (black lines) and O_3 (red lines) measured from the DC-8 aircraft during the TRACE-P transit flight 4 from California to Hawaii and flight 5 from Hawaii to Guam on 25 and 26 February 2001. Profile locations and the flight tracks for flight 4 and flight 5 are shown in Figure 8. Profile A was a double spiral (ascending and then descending) conducted for MOPITT validation to check the consistency of the vertical structure of CO; data from both spirals are shown (there were no observations between 500 and 400 hPa during the ascent). CO measurements between 550 and 300 hPa in profile B are not available. Profiles D and E include descents and ascents separated by 86 km and 275 km, respectively. See color version of this figure at back of this issue.

Table 1. Average Species Concentrations and Ratios Observed in the Trans-Pacific Asian Plume of 26–27 February 2001

	Profile A ^{a,b}	Profile D ^{a,b}	Profile E ^{b,c}
Temperature, °K	253 ± 9	286 ± 1	286 ± 2
CO, ppbv	224 ± 28	198 ± 20	205 ± 29
O ₃ , ppbv	57 ± 1	65 ± 5	74 ± 9
C ₂ H ₂ /CO, pptv/ppbv	3.6 ± 0.2	2.7 ± 0.5	2.9 ± 0.3
C ₃ H ₈ /C ₂ H ₆ , pptv/pptv	0.27 ± 0.05	0.15 ± 0.02	0.14 ± 0.01
C ₂ Cl ₄ /CO, pptv/ppbv	0.041 ± 0.008	0.038 ± 0.013	0.025 ± 0.013
CH ₃ Cl/CO, pptv/ppbv	2.8 ± 0.3	3.1 ± 0.4	3.2 ± 0.3
CH ₃ CN/HCN, pptv/pptv	0.67 ^d	0.73 ^d	0.63 ^d

^aSampled between California and Hawaii on 26 February (average for 600–370 hPa and standard deviation).

^bSee Figure 8 for the locations of the profiles and Figure 9 for the vertical structure of the profiles.

^cSampled between Hawaii and Guam on 27 February (average for 800–600 hPa and standard deviation).

^dLess than three coincident observations.

southern branches. The northern branch continued to North America and was sampled on flight 4 (26 February). The southern branch veered south to the tropical Pacific, subsided and was sampled on flight 5 (27 February). The back trajectories for profiles B and C in Figure 8 suggest some mixing of the Asian plume with tropical marine (B) and upper tropospheric (C) air masses. We limit further analysis to profiles A, D, and E, for which the clusters of back trajectories identify most clearly a common origin.

4.2. Source Characteristics of Trans-Pacific Asian Plume

[24] Both the northern and southern plumes sampled in TRACE-P exhibit the characteristics of recently emitted pollution, as indicated in Table 1. The high C₂H₂:CO concentration ratios (2.7–3.6 pptv/ppbv) are characteristic of fresh Asian outflow [Russo *et al.*, 2003]. The C₃H₈:C₂H₆ ratio for the northern plume is also characteristic of fresh pollution [Russo *et al.*, 2003], but the values for the southern plume are lower and indicate some photochemical aging.

[25] Perchloroethene (C₂Cl₄), a chemical used in dry cleaning, is a tracer of urban pollution whereas methyl chloride (CH₃Cl) is often viewed as a tracer of biomass burning [Blake *et al.*, 2003]. Blake *et al.* [2003] examined the chemical tracer signatures for the high-CO layers in flight 5. They find correlated enhancements of CH₃Cl, C₂Cl₄, ethyne and halocarbons, implying a mixture of fossil fuel and biomass burning (or biofuel) sources. We further examined the CH₃CN/HCN enhancement ratio in an attempt to separate biomass burning from biofuel influence, as this ratio in TRACE-P was much higher in biomass burning plumes (0.46) than in anthropogenic Chinese plumes including biofuel but no biomass burning influence (0.23) [Li *et al.*, 2003; Singh *et al.*, 2003]. We find CH₃CN/HCN ratios > 0.6 in the pollution layers of flights 4 and 5 (Table 1), implying a biomass burning contribution to the Asian outflow. Blake *et al.* [2003] suggest that lower levels of halocarbons and C₂Cl₄ in profile E vs. profile D are indicative of somewhat different origins. We find here that the CH₃Cl/CO ratios were similar in profiles A, D and E, and the C₂Cl₄/CO ratio was consistent within a factor of 2 (Table 1). This together with the back trajectories supports a common origin, however there is clearly some heterogene-

ity present in the plume, which is not inconsistent with WCB lifting air masses from different sources over east Asia.

4.3. Model Simulation and MOPITT Observations

[26] The simulation of this trans-Pacific transport event by GEOS-CHEM is shown in Figure 11. The pollution transport in the model is consistent with the back trajectories shown in Figure 8. The model plume is transported across the Pacific in the midtroposphere (~6 km) and encounters the blocking high-pressure system near the dateline where it splits into southern and northern branches. The southern branch subsides (to 4–5 km altitude) while the northern branch is forced upward (to 7–8 km altitude). The northern enhancement is displaced upward in the model relative to the observed aircraft profiles (Figure 9), which would reduce its sensitivity to observation by MOPITT as discussed in section 3. The tagged tracer simulations indicate that the model enhancement was 70–80% anthropogenic and 20–30% from biomass burning. Such mixing of anthropogenic and biomass burning effluents in the WCBs driving Asian outflow to the Pacific was frequently observed in TRACE-P [Carmichael *et al.*, 2003; Liu *et al.*, 2003; Ma *et al.*, 2003].

[27] Although the model successfully describes the pathways for trans-Pacific transport of the 26–27 February plume, it greatly underestimates (by 50–100 ppbv) the CO maxima observed by the aircraft. Numerical diffusion in the model may be responsible. Such an underestimate was noted previously in the simulation of MOPITT observations of trans-Pacific transport events over the northeast Pacific (Figure 7) and is seen in particular in comparison to the MOPITT column observations for 23–27 February 2001. The daily MOPITT data are relatively sparse, because of incomplete orbit coverage and clouds over the Pacific, but comparison to the model results with averaging kernels applied (Figure 11, center panels) allows one to visualize the Asian outflow plume as it progresses across the Pacific. MOPITT observations indicate high levels of CO reaching North America on 27 February.

4.4. Photochemical Evolution and Ozone Production

[28] Although the pollution layers sampled by the TRACE-P aircraft on flights 4 and 5 originated from the same outflow event from Asia, as confirmed by the back trajectories and the chemical tracer signatures, they show evidence of very different photochemical evolution. We previously commented on the lower C₃H₈:C₂H₆ relationship observed in flight 5. The northern plume sampled in flight 4 exhibited no significant O₃ enhancement (Figure 9, profile A). Conversely, the subsiding southern plume sampled on flight 5 contained large O₃ enhancements (Figure 9, profiles D and E). Differential absorption lidar (DIAL) observations of O₃ vertical cross sections along the tracks of flights 4 and 5 (Figure 12) show no O₃ pollution enhancement at high latitudes (>35°N) consistent with the in situ observations in the northern Asian plume. The DIAL observations show a pronounced high-O₃ layer between 2 and 4 km on flight 5, with levels exceeding 80 ppbv, consistent with the in situ observations of Figure 9 and demonstrating the broad horizontal extent (at least 2500 km) of this subsiding pollution layer. A positive correlation between O₃ and aerosols was also found in these pollution plumes by Browell *et al.* [2003],

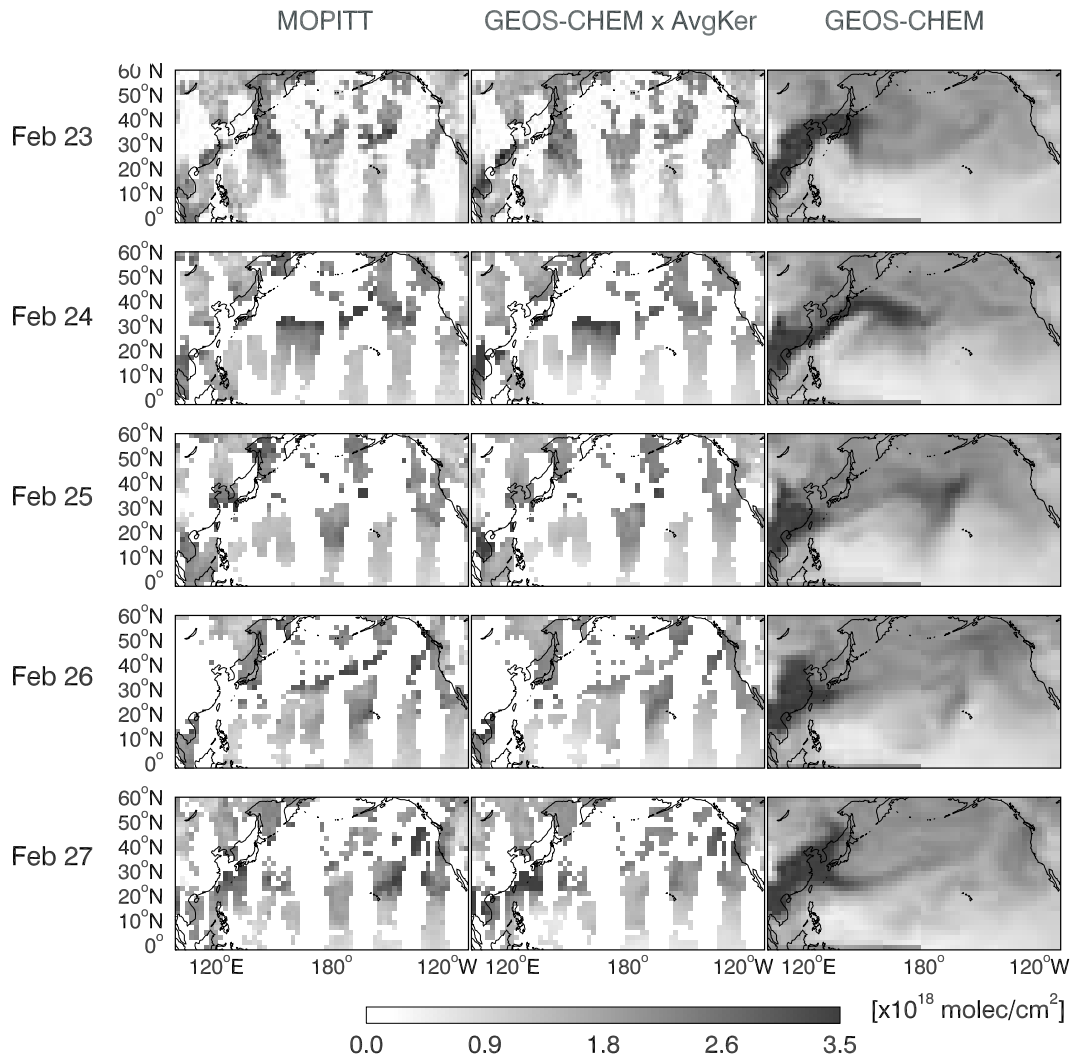


Figure 11. CO columns during the trans-Pacific Asian pollution event on 23–27 February. (left) MOPITT observations. (middle) GEOS-CHEM model results sampled along MOPITT orbit tracks and with averaging kernel applied. (right) Original GEOS-CHEM model results sampled at 1200 UT. The color bar is saturated at 3.5×10^{18} molecules/cm². See color version of this figure at back of this issue.

who attributed the enhancement to biomass burning sources in southeast Asia. The differences in O₃ enhancements between the northern and southern plumes reflects not only the effect of solar radiation but also, and more importantly, the effect of subsidence driving PAN decomposition to NO_x and hence O₃ production. Figure 13 shows the concentrations of NO_y species observed in the northern plume (profile A, flight 4) and in the southern plume (profiles D and E, flight 5). The total NO_y concentration in both plumes is about the same (470–530 pptv), consistent with their common origin as NO_y is conserved. Mean NO_x levels are 18 pptv in the northern plume (4% of total NO_y), and 163 pptv in the southern plume (30% of total NO_y). Total NO_y also includes 13% HNO₃+nitrate aerosol and 74% PAN in the northern plume, vs. 51% HNO₃+nitrate aerosol and 14% PAN in the southern plume (Figure 13). The dominant PAN contribution in the northern plume is consistent with the mean PAN/NO_y molar ratio (0.75 ± 0.10) observed in the warm conveyor belt outflow in the free troposphere during TRACE-P [Miyazaki *et al.*, 2003]. The difference in speciation between the northern and southern plumes can be explained by PAN

decomposition to NO_x during subsidence following the split of the original plume on 25 February. From the temperature measurements in the layers sampled in TRACE-P (Table 1), we find that the PAN lifetime drops from 105 days in profile A to 5–6 hours in profiles D and E.

[29] We can relate the observed PAN decomposition and O₃ production in the southern plume. From Figure 8, O₃ in the southern plume appears to be enhanced by 40 ppbv above the local tropical background. However, considering the midlatitude origin of this plume, a background O₃ concentration of 57 ppbv (such as seen in profile A, Table 1) is more appropriate as a baseline for quantifying the O₃ enhancement in the southern plume (65–74 ppbv, Table 1). We deduce a net ozone production of 8–17 ppbv in the southern plume in the 2 days following the splitting of the plume. Photochemical model calculations by A. J. Hamlin *et al.* (manuscript in preparation, 2003) initialized with the conditions observed in the northern plume yield an O₃ production of only 2–5 ppbv in 2 days over the subsiding trajectory of the southern plume and do not reproduce the observed PAN loss. Increasing the rate of

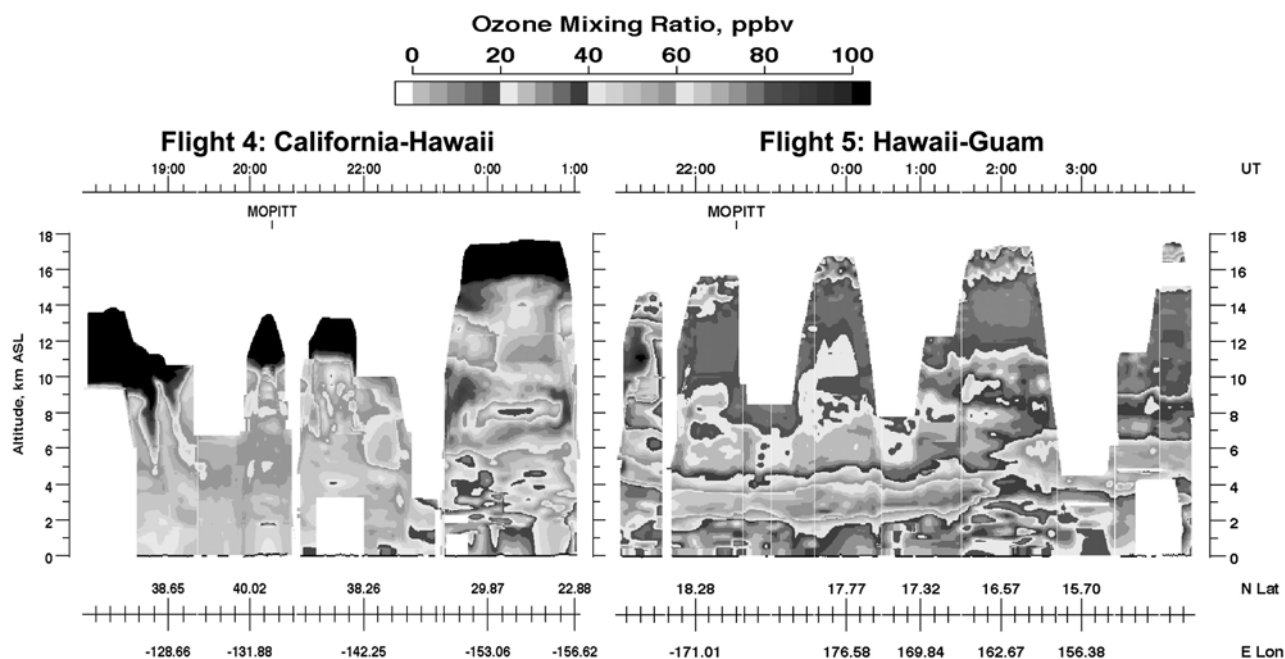


Figure 12. Cross section of O_3 concentrations measured by the differential absorption laser (DIAL) along the tracks of TRACE-P (left) flight 4 between California and Hawaii and (right) flight 5 between Hawaii and Guam. See Figure 8 for details of flight tracks. Extensive Asian pollution layers characterized by elevated CO were sampled by the aircraft between 5 and 10 km altitude on flight 4 and between 2 and 4 km altitude on flight 5. These layers contained elevated O_3 only south of $35^\circ N$, as shown. See color version of this figure at back of this issue.

PAN decomposition by a factor of 2.1–2.3 in that calculation, within the range of the uncertainty in the rate constant [Sander *et al.*, 2003] can match the observed PAN decomposition and produces an additional 2–3 ppbv of O_3 . This O_3 increment matches the O_3 enhancement observed in profile D but still underestimates that in profile E. Interpretation of this discrepancy is difficult as a result of uncertainty in the initial conditions. Heterogeneity within the plume, which was not captured in the model calculations, is a factor of uncertainty in the analysis.

5. Conclusions

[30] We have presented an integrated analysis of continental outflow and trans-Pacific transport of Asian pollution by using aircraft observations from the TRACE-P mission (February–April 2001), concurrent satellite observations of CO columns from the MOPITT instrument, and a global 3-D simulation with the GEOS-CHEM model. We used this analysis to determine the utility of the MOPITT observations to quantify Asian outflow and trans-Pacific transport, and to examine in more detail the sources and photochemical evolution (in particular O_3 production) in a major trans-Pacific event observed during TRACE-P.

[31] The GEOS-CHEM model, previously evaluated with TRACE-P observations of Asian outflow of CO over the northwest Pacific, is found to be highly correlated and globally unbiased with respect to the MOPITT observations during TRACE-P ($R^2 = 0.87$, slope = 0.95). The agreement between MOPITT observations and the GEOS-CHEM model simulation over China suggests no obvious bias in

anthropogenic sources in this region. There are some regional biases, the largest of which is an 18% model overestimate of MOPITT CO columns over Southeast Asia. We estimate that a 60% decrease in seasonal biomass

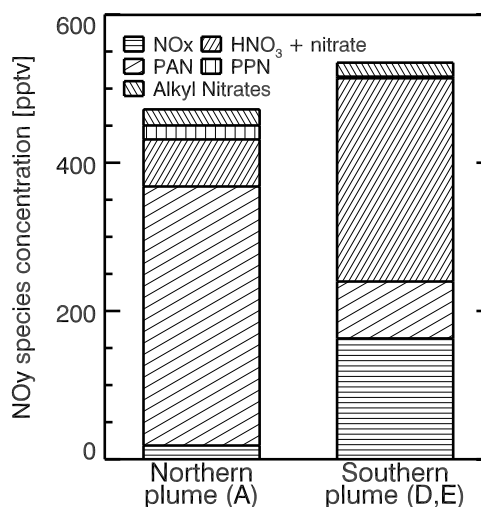


Figure 13. Concentrations of NO_y species in the northern and southern branches of the Asian trans-Pacific plume observed by the TRACE-P DC-8 aircraft on 26 February 2001 during flight 4 between California and Hawaii (profile A between 600–370 hPa) and on 27 February 2001 during flight 5 between Hawaii and Guam (profiles D and E between 800–600 hPa). The concentrations are averages of 8 and 21 observations, respectively.

burning from that region would resolve this discrepancy, and such a correction would be consistent with an inverse model analysis of the TRACE-P CO observations using GEOS-CHEM [Palmer *et al.*, 2003]. An important caveat is that model simulation of the MOPITT column CO is sensitive (through the instrument averaging kernel) to the shape of the CO vertical profile. As an extreme case, suppression of deep convection over Southeast Asia in the model would resolve the regional discrepancy with MOPITT without any change to the biomass burning source. This result highlights the importance of in situ measurements of vertical profiles from aircraft to assist the interpretation of satellite observations.

[32] The MOPITT observations for February–April 2001 identify four major events of trans-Pacific transport of Asian pollution to North America. The GEOS-CHEM model reproduces these events and shows high correlation with the corresponding MOPITT data, degrading however from the northwest to the northeast Pacific. Both numerical diffusion and vertical bias in the model may contribute to the underestimate over the northeast Pacific. All four events observed by MOPITT were also sampled in situ over the northeast Pacific by either the TRACE-P or the PHOBEA-II aircraft missions, or by the Cheeka Peak Observatory in Washington State.

[33] A series of major Asian pollution layers sampled in TRACE-P transit flights from California to Hawaii (26 February) and Hawaii to Guam (27 February) provided an opportunity for a more detailed investigation of the trans-Pacific transport and chemical evolution of these layers, including in particular O₃ production. Concentrations of CO in the layers approached 300 ppbv. Elevated O₃, approaching 80 ppbv, was observed in the low-latitude layers only. Back trajectories and chemical tracers reveal that these layers all originated from a single Asian outflow event on 22 February when a warm conveyor belt ahead of a cold front lifted Asian anthropogenic and biomass burning pollution to the free troposphere. The plume split into southern and northern branches upon encountering a blocking high-pressure system over the mid-Pacific. The MOPITT observations and the GEOS-CHEM model simulation allow a reconstruction of the evolution of this Asian outflow plume during trans-Pacific transport, although GEOS-CHEM does not reproduce the magnitude of the enhancements, perhaps because of numerical diffusion. Elevated O₃ ($\Delta O_3 = 8\text{--}17$ ppbv) was observed in the southern but not in the northern plume. Reactive nitrogen in the northern plume was present mostly as PAN, but subsidence in the southern plume was found to drive PAN decomposition and from there O₃ formation. The ΔO_3 observed in the southern plume was high relative to values obtained from a photochemical model calculation initialized with the conditions of the northern plume. This represents the first field observation of PAN decomposition driving O₃ production in polluted plumes transported to the remote troposphere.

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M. A. Avery, E. V. Browell, J. H. Crawford, G. W. Sachse, and S. A. Vay, NASA Langley Research Center, Hampton, VA 23681, USA. (m.a.avery@larc.nasa.gov; e.v.browell@larc.nasa.gov; j.h.crawford@larc.nasa.gov; g.w.sachse@larc.nasa.gov; s.a.vay@larc.nasa.gov)

D. R. Blake, Department of Chemistry, University of California, Irvine, CA 92697-2025, USA. (drblake@uci.edu)

M. N. Deeter, D. P. Edwards, L. K. Emmons, J. C. Gille, and J. Warner, National Center for Atmospheric Research, Boulder, Colorado, 80303, USA. (mnd@ucar.edu; edwards@ucar.edu; emmons@ucar.edu; gille@ucar.edu; juying@ucar.edu)

A. M. Fiore, C. L. Heald, and D. J. Jacob, Department of Earth and Planetary Sciences, Harvard University, Cambridge, MA 02138, USA. (afiore@fas.harvard.edu; heald@fas.harvard.edu; djacob@fas.harvard.edu)

H. E. Fuelberg, Department of Meteorology, Florida State University, Tallahassee, FL 32306, USA. (fuelberg@met.fsu.edu)

A. J. Hamlin, College of Engineering, Michigan Technological University, Houghton, Michigan 49931-1295, USA. (ahamlin@mtu.edu)

S. T. Sandholm, School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA 30332-0340, USA. (scott.sandholm@eas.gatech.edu)

H. B. Singh, NASA Ames Research Center, MS 245-5, Moffett Field, CA 94035, USA. (hsingh@mail.arc.nasa.gov)

R. W. Talbot, Institute for the Study of Earth, Oceans and Space, University of New Hampshire, Durham, NH 03824–3525, USA. (robert.talbot@unh.edu)

D. J. Westberg, Science Applications International Corp., Hampton, VA 23681, USA. (d.j.westberg@larc.nasa.gov)

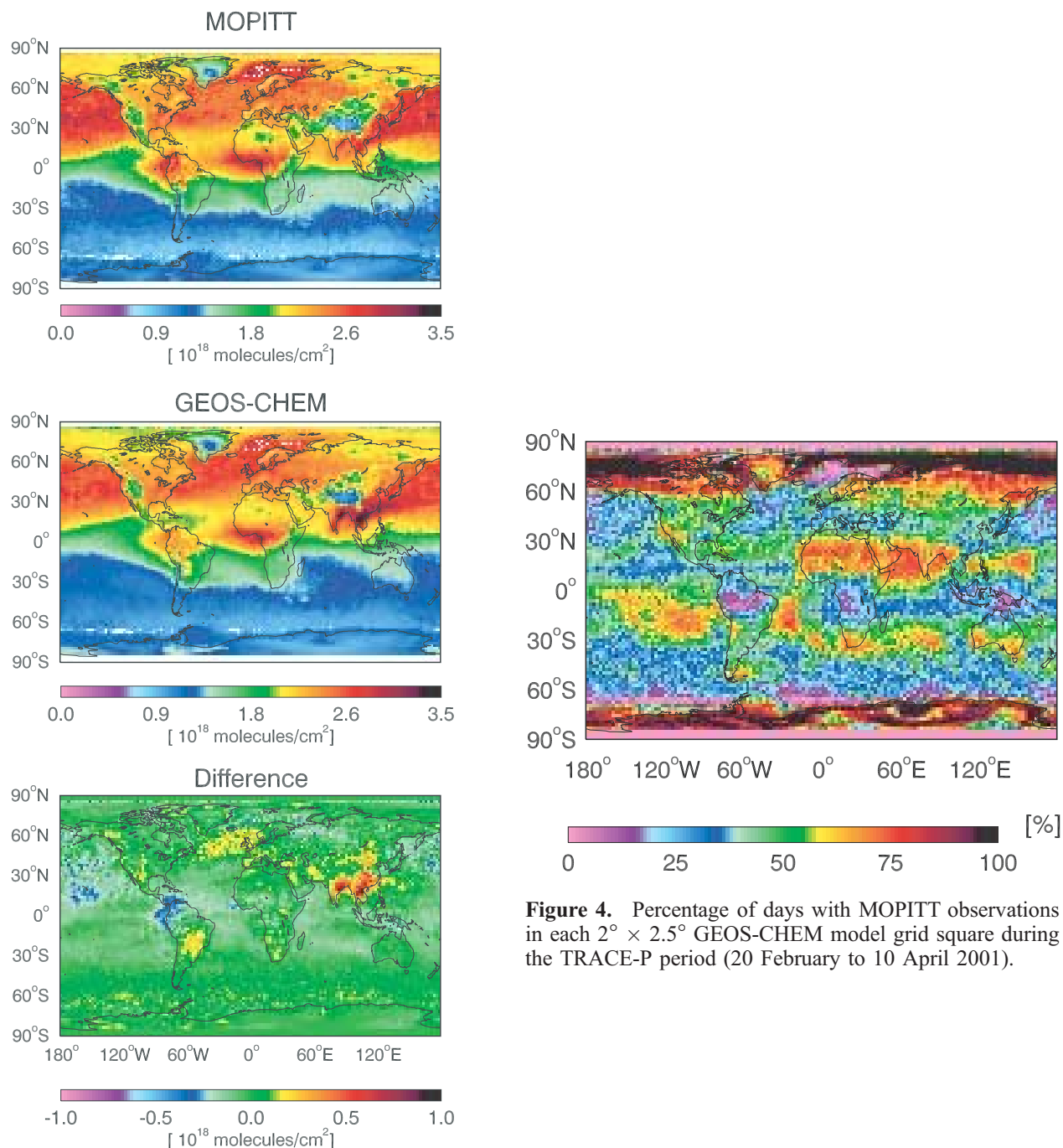


Figure 3. Mean CO columns measured by MOPITT during the Transport and Chemical Evolution Over the Pacific (TRACE-P) campaign (20 February to 10 April 2001) and corresponding GEOS-CHEM model columns sampled along the MOPITT observational track and transformed with the MOPITT averaging kernels. Both the MOPITT data and the transformed GEOS-CHEM fields have been averaged over the GEOS-CHEM $2^\circ \times 2.5^\circ$ model grid. The bottom panel shows the difference between GEOS-CHEM and MOPITT. The color scales for the top two panels are saturated at 3.5×10^{18} molecules/cm², and the bottom panel scale is saturated at 1.0×10^{18} molecules/cm².

Figure 4. Percentage of days with MOPITT observations in each $2^\circ \times 2.5^\circ$ GEOS-CHEM model grid square during the TRACE-P period (20 February to 10 April 2001).

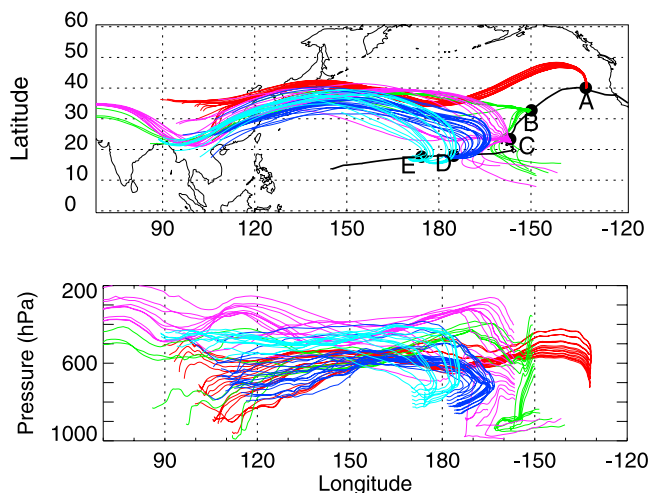


Figure 8. Five-day kinematic back trajectories based on European Centre for Medium-Range Weather Forecasts meteorological fields [Fuelberg *et al.*, 2003] for the enhanced CO layers observed in TRACE-P DC-8 flight 4 from California to Hawaii (26 February) and flight 5 from Hawaii to Guam (27 February). The flight tracks are shown in black, and arrival points in profiles A-E of Figure 9 are shown in red, green, purple, blue, and cyan, respectively.

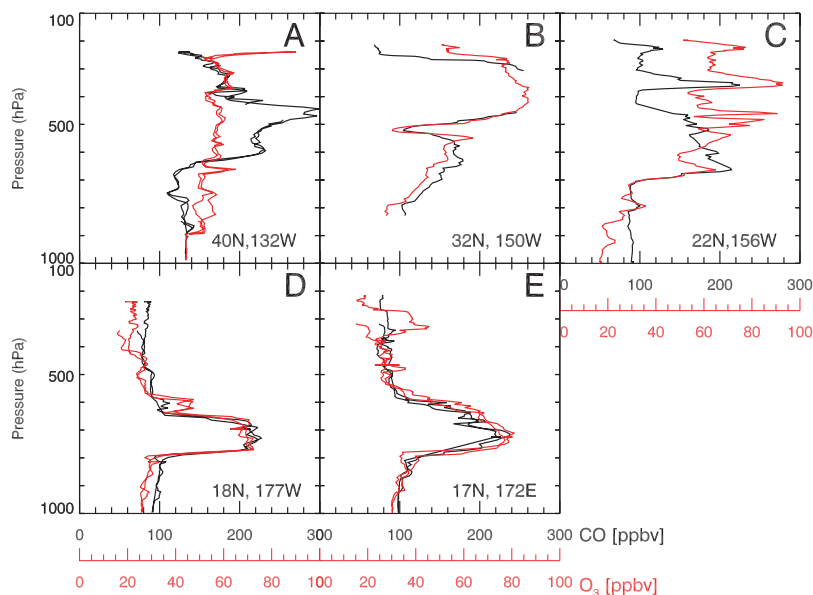


Figure 9. Vertical concentration profiles of CO (black lines) and O₃ (red lines) measured from the DC-8 aircraft during the TRACE-P transit flight 4 from California to Hawaii and flight 5 from Hawaii to Guam on 25 and 26 February 2001. Profile locations and the flight tracks for flight 4 and flight 5 are shown in Figure 8. Profile A was a double spiral (ascending and then descending) conducted for MOPITT validation to check the consistency of the vertical structure of CO; data from both spirals are shown (there were no observations between 500 and 400 hPa during the ascent). CO measurements between 550 and 300 hPa in profile B are not available. Profiles D and E include descents and ascents separated by 86 km and 275 km, respectively.

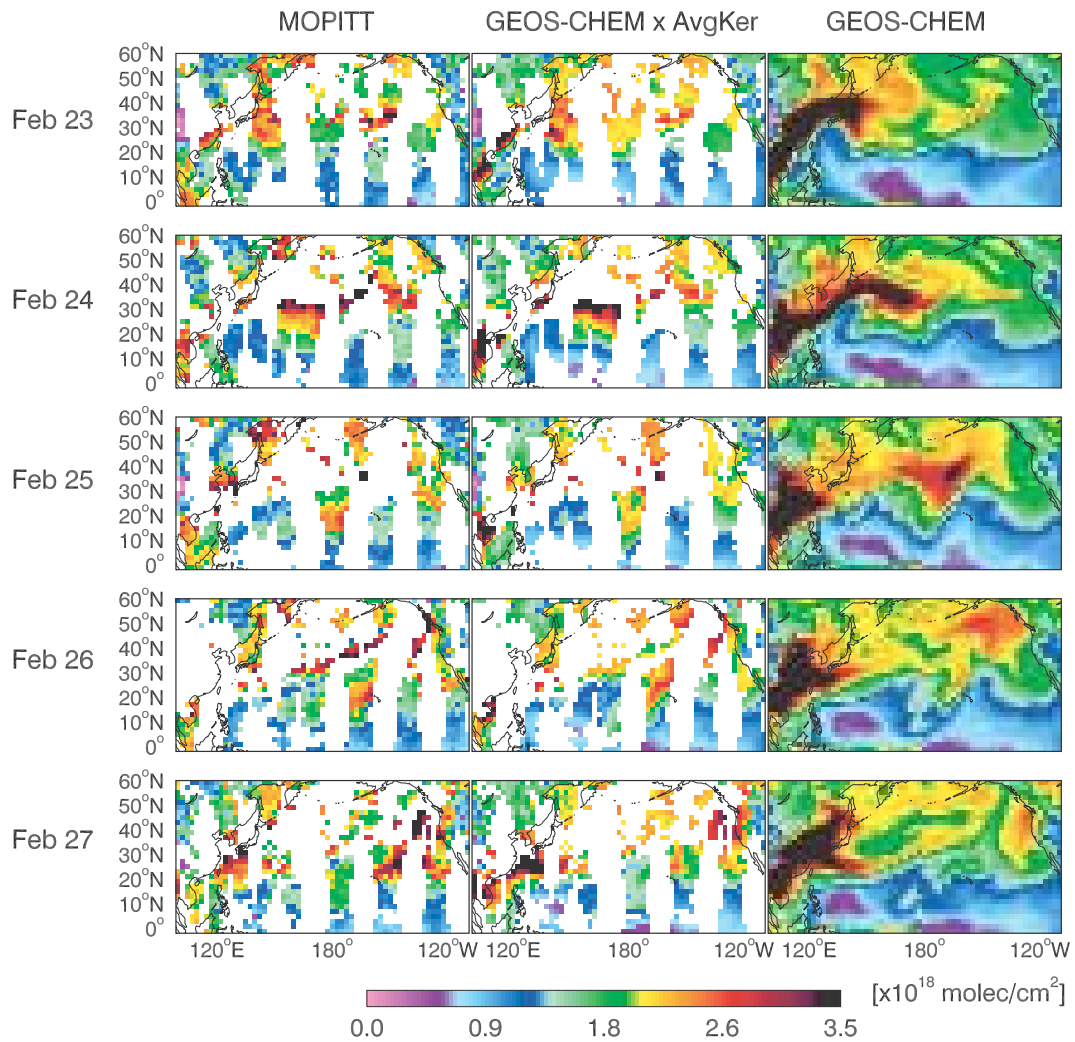


Figure 11. CO columns during the trans-Pacific Asian pollution event on 23–27 February. (left) MOPITT observations. (middle) GEOS-CHEM model results sampled along MOPITT orbit tracks and with averaging kernel applied. (right) Original GEOS-CHEM model results sampled at 1200 UT. The color bar is saturated at 3.5×10^{18} molecules/cm².

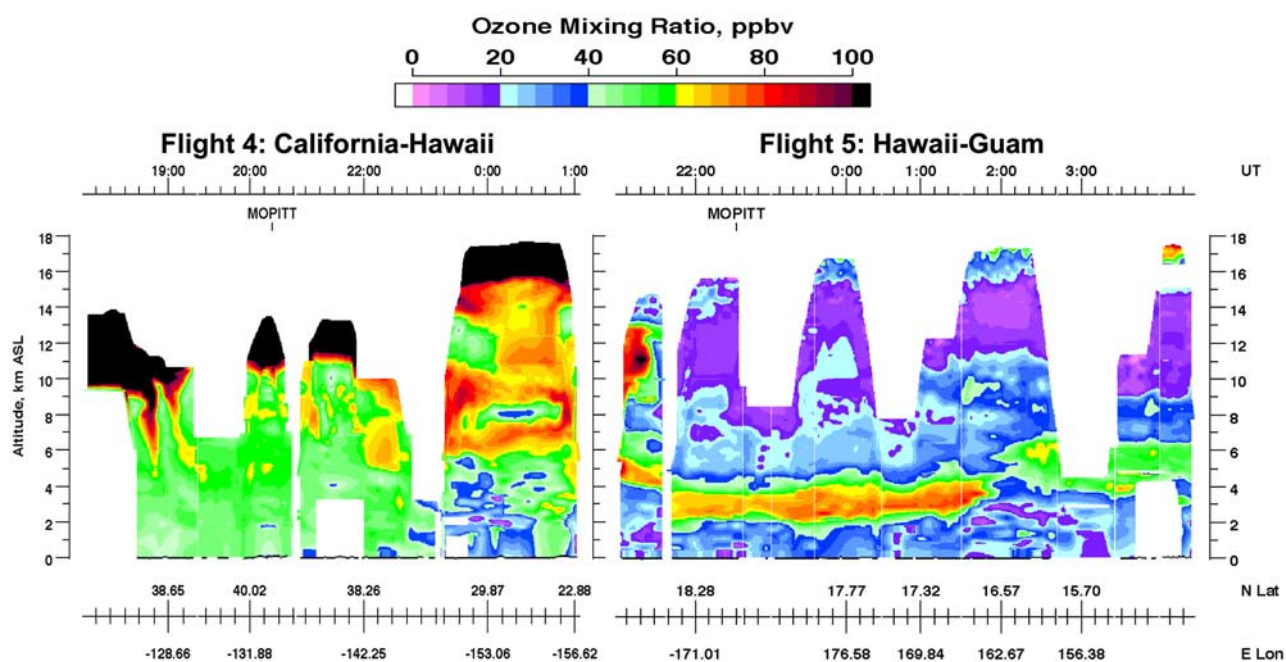


Figure 12. Cross section of O_3 concentrations measured by the differential absorption laser (DIAL) along the tracks of TRACE-P (left) flight 4 between California and Hawaii and (right) flight 5 between Hawaii and Guam. See Figure 8 for details of flight tracks. Extensive Asian pollution layers characterized by elevated CO were sampled by the aircraft between 5 and 10 km altitude on flight 4 and between 2 and 4 km altitude on flight 5. These layers contained elevated O_3 only south of 35°N , as shown.