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Trans-Pacific transport of Asian dust and CO: accumulation of biomass burning CO in the subtropics and dipole structure of transport

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Abstract. In May 2003, both MODIS aerosol optical depth (AOD) and carbon monoxide (CO) measurements from MOPITT show significant trans-Pacific transport to North America. We apply the global chemical transport model, GEOS-Chem, to analyze the main features of the long-range transport events. Enhancements of MOPITT CO over the tropical Pacific are much broader than MODIS AOD enhancements. We find in model simulations that a major fraction of the CO enhancements in the subtropics in May is due to biomass burning in Southeast Asia in April. Biomass burning CO was recirculated into the subtropical high-pressure system and lingered for a much longer period than aerosols transported at higher latitudes. Simulated AOD enhancements are due to a combination of dust, sulfate, and organic and elemental carbons. Dust contribution dominates the AOD enhancements in early May. Model results indicate that dust transport takes place at higher altitude than the other aerosols. MODIS observations indicate a bias in model simulated pathway of dust transport in one out of the three cases analyzed. Sensitivities of dust transport pathways are analyzed in the model. The dipole structure of transport, consisting of the Aleutian Low to the north and the Pacific High to the south, over the Pacific is found to be a key factor. The placement of the dipole structure relative to model parameters such as up-stream wind field and source location may lead to the high sensitivity of simulated transport pathways.

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1 Introduction

Trans-Pacific transport of Asian pollutants is evident in in-situ and satellite observations (Merrill et al., 1989; Parrish et al., 1992, 2007; Jaffe et al., 1999; Wang et al., 2003, 2006). Global model simulations showed that the transport has an impact on regional air quality as well as radiative forcing in the United States (Jacob et al., 1999; Takemura et al., 2002; Chin et al., 2002; Park et al., 2003; Hadley et al., 2007; McNaughton et al., 2009; Leaitch et al., 2009). The transport events of pollutants, affecting the contiguous United States, largely occur on a continuous basis throughout the year (Liang et al., 2004), while the transport events of dust occur on average once a year, preferentially in spring, with intensive occurrences in just selected years (1993, 1998, 1999 and 2001) (Jaffe et al., 1999; Clarke et al., 2001; Jaffe et al., 2003; Heald et al., 2006). These events lead to episodic increases in atmospheric concentrations of dust and pollutants in the downstream regions (Parrish et al., 2007; Leaitch et al., 2009) or year round enhancements in the background levels, particularly in the west coast of the United States due to Asian pollution transport (VanCuren et al., 2005; Hadley et al., 2007). A typical transport event takes approximately 6–8 days across the Pacific (Takemura et al., 2002; Stohl et al., 2002; Holzer et al., 2003; Leaitch et al., 2009).

A variety of studies have been conducted for better understanding of Asian pollutant transport and they have provided a lot of insights into the impacts of the transport. However, there still remain large uncertainties in model calculations (e.g., Chin et al., 2002). Specifically, the state of the art models are capable of simulating the approximate transport pattern but are not suitable for quantitative analysis in some cases (Heald et al., 2006; Dunlea et al., 2009). Due to the general lack of observations, only a limited number of trans-Pacific transport events have been used in model evaluations

and characterizations of the transport events. Over the vast stretch of the Pacific Ocean, satellites are often the only observation platform available. The advantage of satellite observations is that multiple species are available with extended spatial coverage for multiple years.

As part of the NASA Earth Observing System (EOS) program, the Terra and Aqua satellites were launched in 1999 and 2002, respectively. They both have sun-synchronous, near-polar, and circular orbit. Aboard both satellites, the MODerate resolution Imaging Spectroradiometer (MODIS) (Salomonson et al., 1989; King et al., 1997) monitors the globe with a wide spectral range, with near daily global coverage and relatively fine spatial resolution. The Aqua satellite passes across the equator in an ascending node at approximately 13:30 local time, while the Terra satellite passes across the equator in a descending node at 10:30. The difference in orbits results in different view and cloud conditions for observations at a given location. Hence MODIS AOD measurements from both platforms were analyzed in this study. The aerosol algorithm of MODIS has been validated under a variety of conditions (Remer et al., 2002; Levy et al., 2003; Chu et al., 2005). Along with the aerosol measurements from MODIS, we used carbon monoxide (CO) measurements from the Measurement Of Pollution In The Troposphere (MOPITT) (Clerbaux et al., 2002; Deeter et al., 2002; Emmons et al., 2008) in this study. MOPITT aboard the Terra satellite was designed to measure atmospheric concentrations of a number of gases and meteorological variables.

Relative to in situ observations, satellite observations from MODIS and MOPITT provide limited or no information in the vertical distribution in the troposphere and their retrieval uncertainties are larger. We attempt to minimize the uncertainty of the measurements by finding the strongest trans-Pacific transport events in the satellite observations and apply a global chemical transport model (CTM), GEOS-Chem, to analyze these events. In the analysis, we take into consideration the measurement uncertainties and focus the modeling analysis on the characteristics well defined by the satellite measurements. In this study, we scanned the 2002-2008 AOD measurements from MODIS to identify strongest trans-Pacific transport events of Asian aerosols. MODIS AOD measurements have larger uncertainties than MOIPITT CO, and therefore corresponding CO measurements from MOPITT were subsequently examined to confirm the transport events. May 2003 was found to be the period of largest enhancements of trans-Pacific aerosols as well as CO through visual inspection of the 7-year data. Three events were identified for this month. We applied the GEOS-Chem model to simulate the long-range transports of aerosols and CO across the Pacific in these events. We find two major issues in the model simulations of trans-Pacific CO and dust enhancements in one out of the three cases. We then examine the physical processes represented in the model to understand the mechanisms.

2 Model description

We use the GEOS-Chem global 3-D model (Bey et al., 2001a) to simulate the trans-Pacific transport of aerosols and CO in May 2003. The model (version 7.3.6) is driven by assimilated meteorological fields of the Goddard Earth Observing System (GEOS) version 4 and contains a detailed oxidant-aerosol chemical mechanism. The GEOS-Chem model we used in this work calculates concentrations of aerosol and gaseous trace species at a horizontal resolution of $2^\circ \times 2.5^\circ$ for 30 vertical layers, 17 of which are in the troposphere. Within the model, aerosols are assumed to be externally mixed and the AOD at 550 nm for each aerosol component is calculated using the formulation by Tegen and Lacis (1996). We conducted a simulation of aerosols and CO for a 7-month period, including a 6-month model spin-up and a 1-month simulation for the analysis of the transport events.

The simulations of aerosols and CO in this study are similar to the previous study by Heald et al. (2006) with minor modifications as described below. The global sources of sulfur, ammonia, and NO_x are described by Park et al. (2004). Carbonaceous aerosol emissions are taken from Cooke et al. (1999) for fossil fuel sources and Yevich and Logan (2003) for biofuels. Sea salt and dust emission are calculated following the schemes of Monahan et al. (1986) and Ginoux et al. (2001), respectively. We employed the dust mobilization scheme by Zender et al. (2003) in place of that by Ginoux et al. (2001) to test the sensitivity of uplifting of dust particles and subsequent transport to dust emission schemes. For both simulations, GOCART source function (Ginoux et al., 2001) was used. Implementation of dust emission and mobilization schemes (Ginoux et al., 2001; Zender et al., 2003) in GEOS-Chem is described by Fairlie et al. (2007). Details of the standard GEOS-Chem formulation can be found elsewhere (Park et al., 2003, 2004; Heald et al., 2006, and references therein).

We calculated contribution of anthropogenic and biomass burning sources in Asia to the column CO distributions over the Pacific using tagged CO tracers (Bey et al., 2001b) by source type and region. The tagged CO simulation was initialized through the 6-month model spin-up as the standard full chemistry simulation, and it used monthly mean OH concentration field archived from a previous standard simulation of year 2001 by Evans et al. (2005).

Biomass burning emissions of Elemental Carbon (EC), Organic Carbon (OC), and CO are based on the dry mass burned data with an averaging period of 8 days from the Global Fire Emission Dataset (GFED) (Randerson et al., 2008). Year-specific biomass burning emissions were used for both full chemistry and tagged CO simulation due to the significant inter-annual variability as shown in the previous studies (Park et al., 2003; Chin et al., 2007). Since this study is focused on events with a duration of about a week, emission estimates with a finer temporal resolution, than the monthly averages typically used, are used. Global

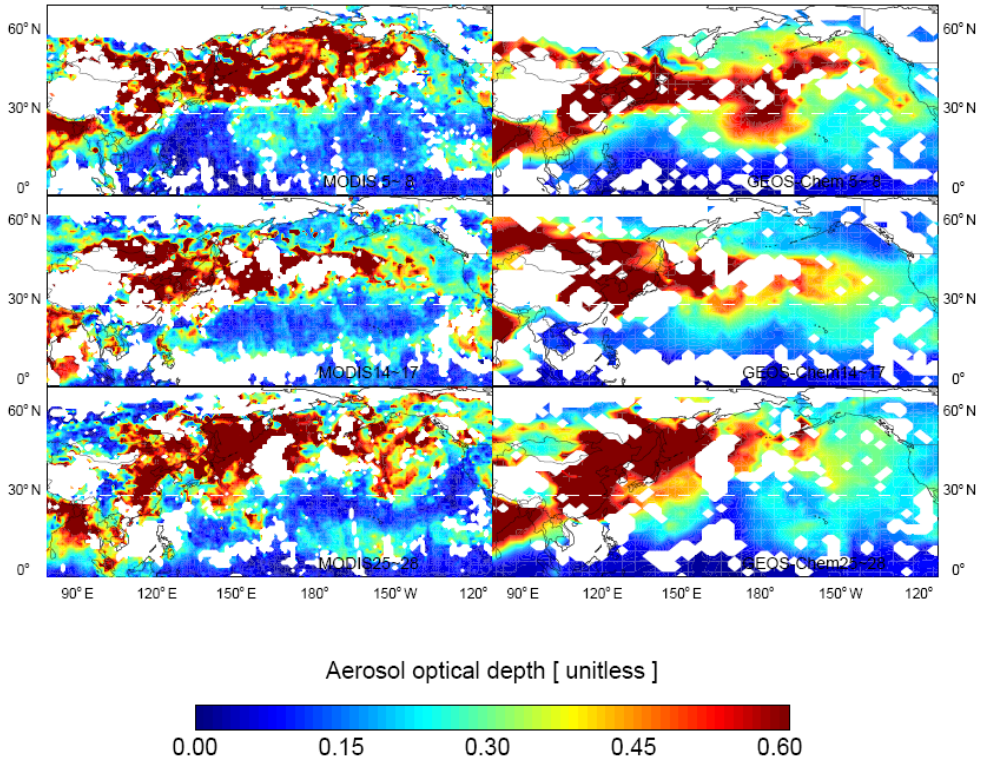

Fig. 1. AOD at 550 nm over the Pacific during 5–8 (first row), 14–17 (second row), and 25–28 (third row) of May 2003 observed by MODIS (first column) and simulated by GEOS-Chem (second column).

emissions from wildfires of EC, OC, and CO are in the ranges of 0.03–0.1 Tg C, 0.23–1.12 Tg C, and 4.48–16.36 Tg CO, respectively, for each 8-day period in spring 2003. The variation of emissions within a month is up to 35% from the monthly average values.

3 Spatial patterns of trans-Pacific transport events

MODIS AOD measurements from both the Terra and the Aqua indicate a series of strong trans-Pacific transport events of Asian aerosols in May 2003. Figure 1 compares observed and simulated AOD during three events, on 1–8, 10–17, and 21–28 May 2003. Only Aqua observations are shown, since Terra MODIS shows comparable results for all the events. MODIS AOD measurements indicate approximately an 8-day duration of individual transport events. Here, we present average values for the last 4 days of the transport events, in which AOD enhancements due to trans-Pacific transport are well formed. This averaging compromises between MODIS sparse daily spatial coverage (due mainly to cloud interference) and the deformation of transport plumes over time. Only GEOS-Chem results, corresponding to the clear-sky MODIS measurements, were used for averaging. It is apparent that the model AOD values are lower than MODIS retrievals over the Pacific. The previous study by Heald et al. (2006) also reported GEOS-Chem underestimation of AOD over the Pacific during a period of massive dust storms in April 2001. The model shows a low bias with respect to MODIS, which is likely due to a known high bias for MODIS in the presence of dust (Levy et al., 2003; Chu et al., 2005). Quantitative assessments of the simulated AOD values are therefore severely limited by the retrieval uncertainties, therefore we focus the analysis on the transport pathway patterns.

The MODIS measurements show slightly different transport pathways in three transport events; the plume in the second event was transported at lower latitudes across the Pacific than the first and third events. The transport pathway patterns were simulated reasonably well in the second and third events, while the simulated plume in the first event is clearly shifted to lower latitudes compared to the observations, resulting in model underestimates of aerosol loading in the North Pacific and overestimates in the Central Pacific.

4 Accumulation of CO over the Central Pacific

Before going into detailed model analysis of AOD simulations, we first evaluate model simulated column CO with MOPITT observations (Fig. 2). The sensitivities of MOPITT to tropospheric CO have altitude dependence (Deeter et al.,

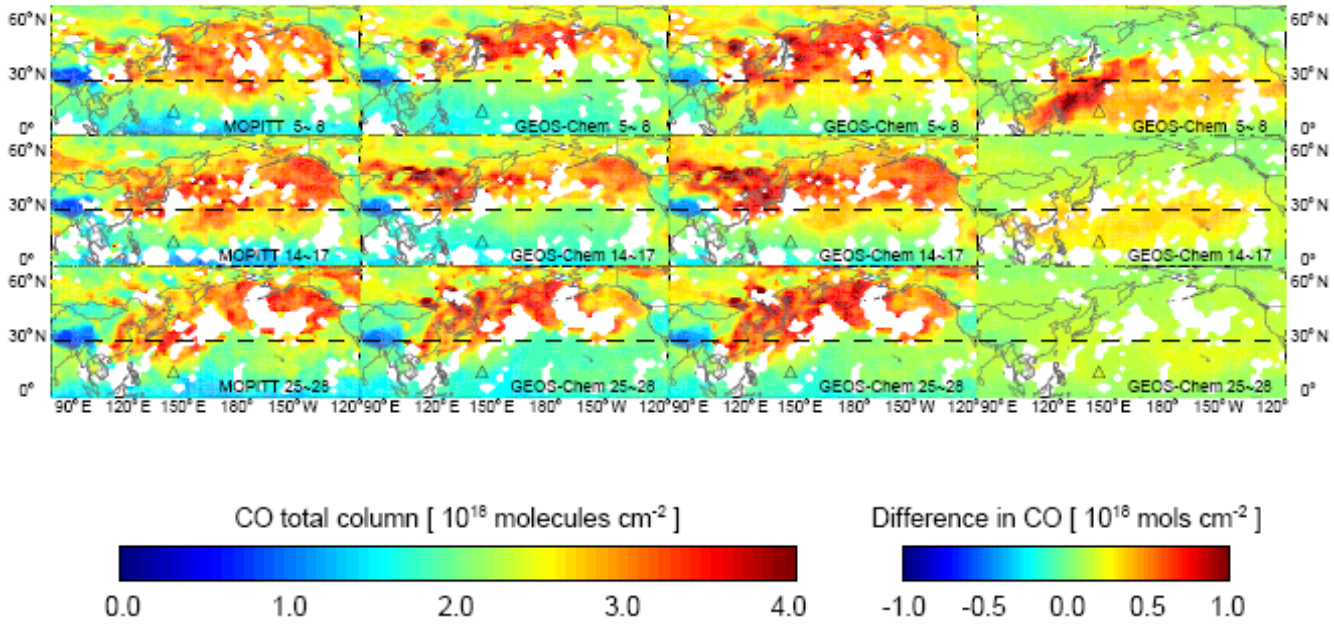

Fig. 2. CO total columns over the Pacific during 5–8 (first row), 14–17 (second row), and 25–28 (third row) of May 2003 observed by MOPITT (first column) and simulated by GEOS-Chem with original biomass burning emissions (second column) and with enlarged biomass burning emissions (third column). Indochina biomass burning emissions in April (not May) are increased by a factor 8 in this simulation. The fourth column shows the difference of column CO in this simulation (third column) from the standard simulation (second column). MOPITT averaging kernel was applied to the model results. The location of the NOAA ESRL monitoring site (GMI) is shown as an open triangle.

2002). We process the model results with MOPITT averaging kernel for a proper comparison. A careful inspection of the observed and simulated column reveals that while the model generally captures the observed magnitude and distribution of MOPITT CO, the observed high CO columns extends further south to the Central Pacific than the model results.

The dichotomy of good simulation of the northern part of the plumes but significant underestimation of the southern part of the plumes is likely driven by an underestimation of CO emissions at lower subtropical latitudes. Heald et al. (2003) indicated that biomass burning effluents of southeast Asia are transported over the Pacific at lower latitudes than Asian anthropogenic pollution. According to the GFED inventory, biomass burning released a total of 3.58 and 0.11 Tg CO during April and May 2003, respectively, in the Indochina Peninsula (Fig. S1 in the electronic supplement, http://www.atmos-chem-phys.net/10/3297/2010/acp-10-3297-2010-supplement.pdf). We increased the CO emissions in that region by a factor of 2, 4, and 8, respectively to investigate the sensitivity of column CO over the Central Pacific to this emission. Figure 2 shows the model results when Burma biomass burning was increased by a factor of 8. Column CO is enhanced more over the Central Pacific than the North Pacific. While a factor of 8 increase leads to some overestimates of column CO compared to MOPITT, a factor of 4 increases underestimates, suggesting a factor of 4–8 increases of emissions in the region based on MOPITT observations.

The underestimation of the biomass burning source in southeast Asia was corroborated by the tagged CO simulation (Figure S2 in the electronic supplement, http://www.atmos-chem-phys.net/10/3297/2010/acp-10-3297-2010-supplement.pdf). While the contribution of biomass burning CO from southeast Asia is mainly over the subtropical Pacific Ocean, the contribution of anthropogenic emissions from southeast Asia produce a large latitudinal gradient from the subtropics to mid and high latitudes. Since the model does not underestimate CO columns at mid and high latitudes (Figure 2), it is more likely that the emission bias is from biomass burning than anthropogenic emissions. Biomass burning in the Indochina Peninsula occurred mostly in April 2003, and −70% of the CO emissions occurred in the first half of April. We show the effects of increasing biomass burning by a factor of 8 only in April in Fig. 2. Due to its long lifetime against oxidation, CO emitted in April 2003 could affect the atmospheric distribution of CO one month later. Biomass burning CO from the Indochina Peninsula in April was recirculated into the subtropical high-pressure systems, subsequently transported to the Central Pacific, and lingered towards the end of May. The CO enhancements decrease as CO is

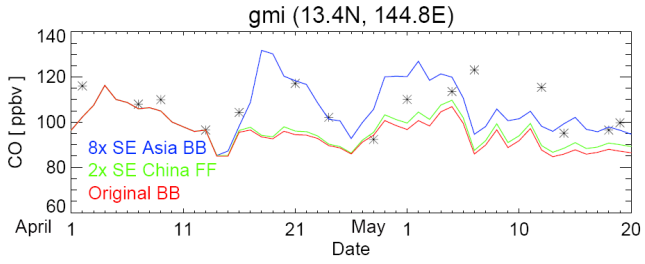

Fig. 3. Time series of simulated (lines) and observed (asterisks) CO mixing ratios at the Mariana Island site in Guam from 1 April to 20 May 2003. The model result using the GFED CO inventory is shown in red line. The sensitivity result with 8 times biomass burning CO emissions from the Indochina Peninsula (only in April) is shown in blue line.

oxidized. We see in Fig. 2 the large effect in the first event and more moderate effects in the second and third events.

To analyze further the impacts of Southeast Asian biomass burning emissions, we analyze surface observations by NOAA ESRL (Fig. 3, Novelli et al., 2008). Among the active ESRL sites in April–May 2003, the Mariana Island site in Guam (GMI, the location of which is shown in Fig. 2) is in the closest vicinity of the biomass burning plumes (Fig. 2). While it is further south from the plume pathways, Fig. 2 shows that the site is located in a region sensitive to biomass burning emissions from the Indochina Peninsula. When biomass burning emissions from the Indochina Peninsula are increased by a factor of 8 in April only, the model result captures the observed CO enhancements. The CO increase relative to the original GFED inventory is 41% in the middle of April and up to 26% in the beginning of May. Although 70% of biomass burning emissions from the Indochina occurred in the first half of April in the GFED inventory (Fig. S1 in the electronic supplement, http://www.atmos-chem-phys.net/10/3297/2010/acp-10-3297-2010-supplement.pdf), the simulated CO enhancements due to increase of biomass burning emissions become significant in the second half of April, reflecting in part the relatively slow transport in the subtropics compared to mid and high latitudes in April and May (winds at 500 and 900 hPa will be shown in Fig. 8). We conduct another sensitivity simulation, in which the anthropogenic CO emissions over southeast China are doubled. Since the CO enhancements over the subtropical Pacific are only apparent in the first half of May (Fig. 2), the doubling of CO emissions is applied in the 6 months before May. The effects on surface CO at the GMI site and the distributions of CO over the Pacific are relatively minor (Fig. 3 and Fig. S3 in the electronic supplement, http://www.atmos-chem-phys.net/10/3297/2010/acp-10-3297-2010-supplement.pdf).

The simulated CO enhancement from increased biomass burning lasted into the first week of May (Fig. 3). Simulated CO is lower than the observation on 12 May but is in agreement on 14 May, indicating that either biomass burning CO in Southeast Asia is underestimated in early May or that the accumulation of biomass burning CO from April dissipates too fast in the model because of model transport error. Additional measurements between 6 May and 12 May would be necessary for more detailed modeling analysis.

5 Characteristics of transport events

We use the GEOS-Chem results to apportion AOD into dust, sulfate, carbonaceous (EC and OC), and sea salt aerosols in the troposphere. Figure 4 compares the trans-Pacific distribution of dust AOD with all the other aerosol AOD (total AOD – dust AOD) simulated by the model. Among the three events, dust AOD contributes as much and more to the total AOD over central and eastern Pacific than the other aerosols during the first event. It is also the event when dust AOD is transported further into the West Coast of the United States than the other aerosols. In the other two events, dust AOD tends to lag behind the other aerosol AOD. The latitudinal displacement of transport pathways between dust and the other aerosols is also most significant in the first event. The large dust signal in the first event provides an opportunity to use MODIS measurements to assess dust transport. Sulfate aerosols account for 40–60% of all the other aerosol AOD (except dust) over the Pacific; carbonaceous and sea salt aerosols account for the rest. Dust and sulfate aerosols together contribute 70–80% of the total AOD downwind during the first and the second events and 50–60% during the third event. The relative contribution from carbonaceous aerosols in the third event is larger than the two other events.

In addition to horizontal distribution, we examine the altitude distribution of aerosol transport simulated by the model (Fig. 5). In all three events, dust is transported at higher altitudes, mainly 500 hPa, than the other aerosols and CO. For this simulation, the standard biomass burning CO inventory without increased emissions in Burma area described in the previous section was used. It is transported faster since the strength of westerlies increases in altitude. Transport of the other aerosols and CO occurred at relatively low altitudes, extending from ground up to 600 hPa. Not only was the transport slower at low altitude, as shown in the longitudinal extent of AOD, the horizontal distributions of column AOD are also different (Fig. 4) reflecting in part the vertical wind sheer in the mid-latitude westerlies over the Pacific. Simulations using the dust mobilization scheme by Zender et al. (2003) show similar results in both horizontal and vertical distributions (not shown). Since sulfate and hydrophilic organic aerosols can be easily scavenged during convection, our selection of largest MODIS aerosol AOD enhancement events over the Pacific may have led to a study period with fewer convections than normal over East China. As a result, the pollutant transport from East China is mainly at low altitudes.

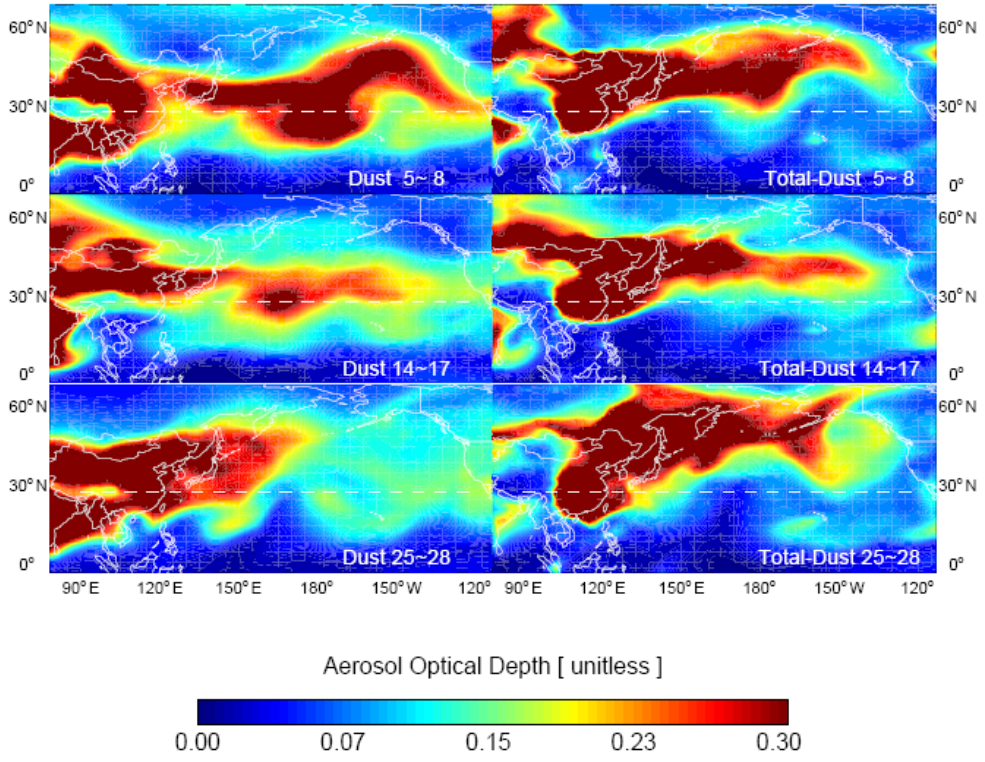

Fig. 4. AOD at 550 nm of dust (left) and of all the other aerosols (right) simulated by GEOS-Chem over the Pacific during 5–8 (first row), 14–17 (second row), and 25–28 (third row) May 2003.

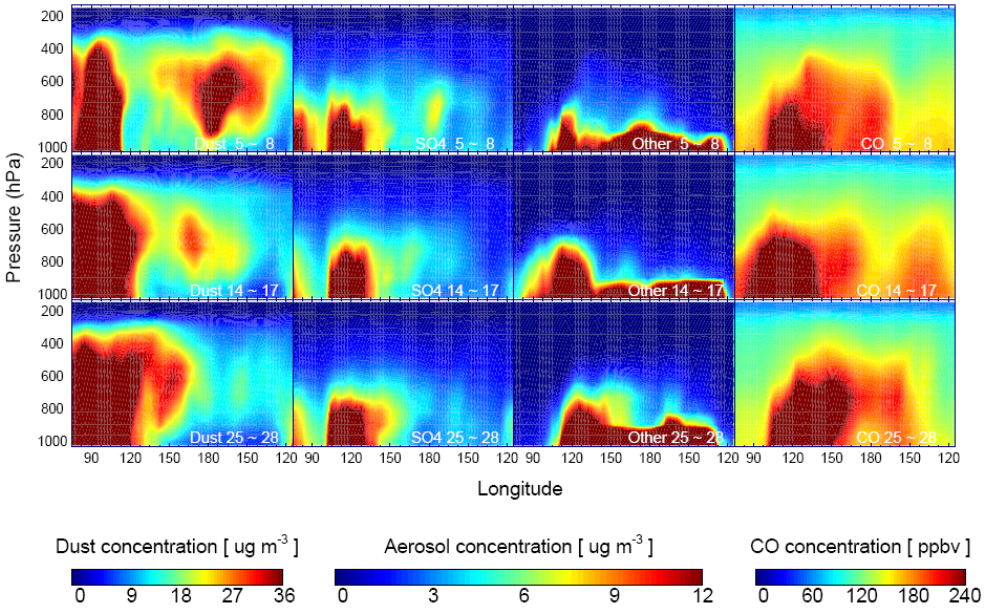

Fig. 5. Cross sections of simulated mixing ratio of dust (first column), sulfate (second column), and the rest of aerosols (third column), and simulated CO mixing ratio during 5–8 (first row), 14–17 (second row), and 25–28 (third row) May 2003 across the Pacific, averaged over 40–60° N as a function of longitude and altitude.

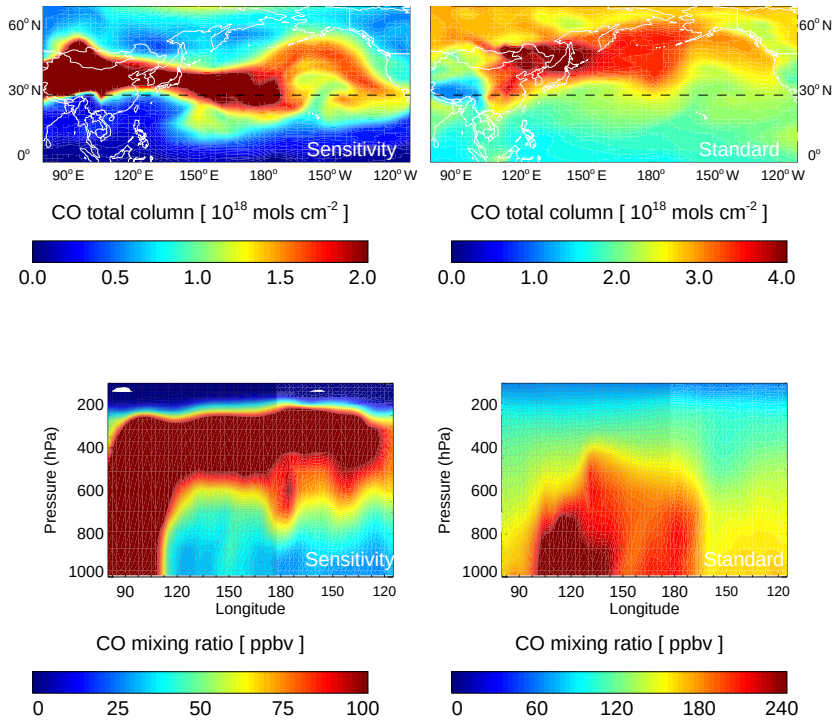

Fig. 6. Horizontal (first row) and vertical (second row) distributions of CO transport in the sensitivity (first column) and standard simulations (second column). The GFED biomass burning emissions are used in both simulations. See text for the details of the sensitivity simulation.

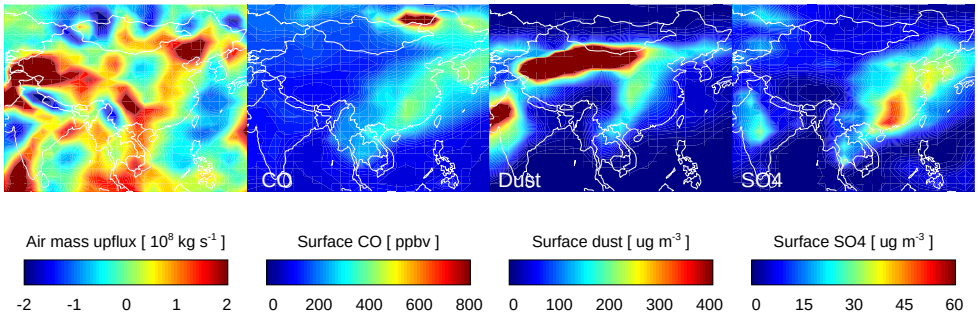

Fig. 7. Distributions of tropospheric vertical air mass flux, and simulated CO, dust and sulfate mixing ratio in the first model layer during 1–2 May 2003.

While the transport pathways of simulated AOD is veered southward compared to the observations (Fig. 1), transport pathway of CO simulated using the biomass burning inventory with increased emissions in Burma area is consistent with observations (Fig. 2). These comparisons indicate that the bias in AOD transport is driven by the bias of dust transport (Fig. 4). A key reason for the large difference in the simulated transport pathways of dust and the other aerosols is the geographical distributions of their sources. Dust sources are located over the two most active deserts in Asia, the Taklimakan and Gobi deserts, in the model. The other aerosols are emitted mainly from anthropogenic sources over East China and biomass burning over southeastern Siberia along with CO. Meteorological transport for dust therefore differs from the other aerosols or CO in the model. We explore in detail the factors contributing to the model bias in dust transport pathways during the first event.

6 Model bias in dust transport pathway during the first event

We chose the first event since the dust signal and model bias are larger than the other two events. We first conduct a sensitivity simulation of CO because its chemical lifetime is long

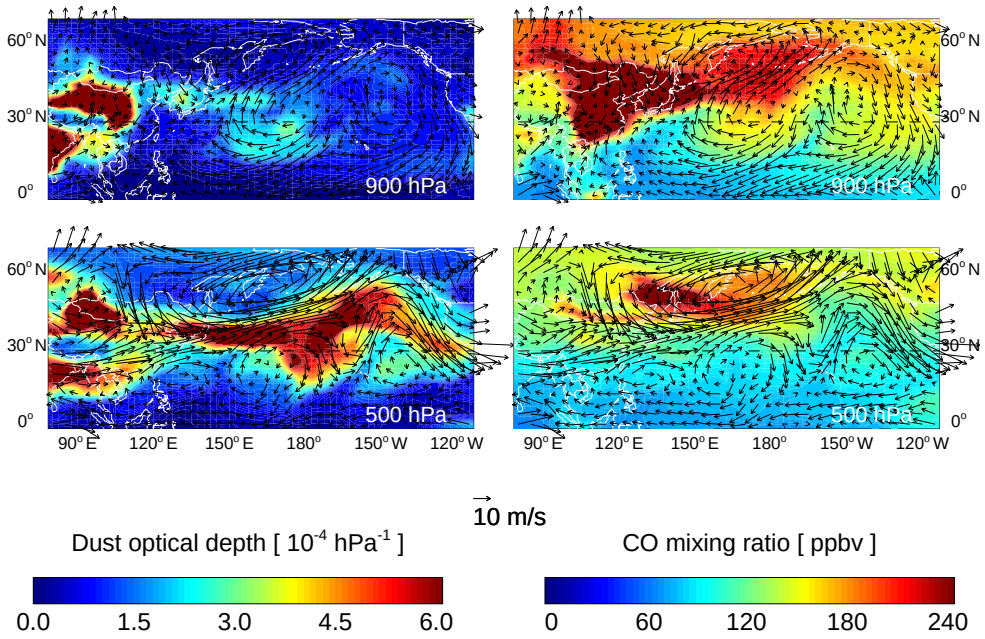

Fig. 8. Wind vectors at 900 hPa (first row) and 500 hPa (second row) used for model simulation along with the spatial distributions of dust (first column) and CO (second column) during 5–8 May 2003 over the Pacific. The GFED biomass burning emissions are used.

and model simulated transport pathway of CO (using an inventory with increased biomass burning emissions in Burma) is consistent with the observations (Fig. 2). In the sensitivity simulation, we redistribute CO emissions for East Asia (∼125 Tg CO) from a month before the first event onset (30 March) through the end of the first event (8 May) evenly in the dust source regions (to be shown in Fig. 7).

Simulated horizontal and vertical distributions of CO transport are compared between the sensitivity and the standard simulations in Fig. 6. In the sensitivity simulation, transport of CO released in dust source regions is veered southward to lower latitudes and at higher altitudes (300–500 hPa) than in the standard simulation. The transport pattern is consistent with transport of dust shown in Figs. 4 and 5. The uplifting of dust in the standard model is therefore related to meteorological conditions near the source regions.

The meteorological fields used for model calculation were analyzed for its contribution to the model bias. In the following analysis, we show the meteorological fields in the first 2 days of the event if transport of interest is near the source regions and the meteorological fields in the second half of the event if transport is over the Pacific. The period of dust event was identified based on MOIDS AOD measurements (not shown). Figure 7 shows the distribution of averaged tropospheric vertical air mass flux over the period of the first 2 days (1–2 May) of the event. Large areas of upward flux are found over the dusty regions north of the Tibetan Plateau. In contrast, high surface CO and sulfate are located in regions with weak uplifting. Furthermore, in close proximity to dust source region, strong upward flux is found throughout the first two days of the event. Once lifted into the upper troposphere, dust is transported quickly by strong westerlies.

It is possible that vertical transport is overestimated by the model over the dust source regions. We therefore conduct a second sensitivity simulation, in which vertical advection of dust is reduced by 50%. Suppressing vertical transport did not affect the spatial distribution of trans-Pacific dust transport (not shown). A third sensitivity study was conducted to investigate the impact of deep convection on dust lifting. Deep convection did not play a significant role in lifting dust particles (not shown), as we would not expect deep convection over arid regions. Therefore, the transport appears to be more sensitive to the location of sources than the rate of uplifting.

Figure 8 shows horizontal wind vectors in two different vertical layers (900 and 500 hPa) and the corresponding distributions of dust and CO during the second half of the first transport event. The divergence of transport over the Pacific is driven largely by a dipole structure of clockwise high-pressure system (the Pacific High) to the south and counter-clockwise low-pressure system (the Aleutian Low) to the north (also noted by Holzer et al., 2005; Liang et al., 2005; Liu et al., 2005; Zhang et al., 2008). The strongest transport is between the two systems, where the pressure gradient is largest. Because of the difference in emission locations, simulated CO transport is mostly caught by the northern counter-clockwise transport towards the northeast, whereas the simulated dust transport is mostly caught by

the clockwise transport towards lower latitudes. Southward transport by the Pacific High is even stronger at higher altitude where dust transport mostly occurs. The transport pathway divergence due to the dipole structure of atmospheric circulation may be common in late spring. Previously, Wang and Zeng (2004) showed the model transport error in May 2000 diagnosed by the correlations of ethane and propane.

Figures 1 and 4 clearly indicate that the simulated dust transport is veered too far south in the model. We suggest here two possible reasons for the model bias. First, it may be related to a potential model bias of (horizontal) advection. We compare the GEOS-4 wind fields at 500 hPa and 900 hPa with the reanalysis from the National Centers for Environmental Prediction (NCEP) (Kanamitsu et al., 2002) in the first two days of the event (Fig. S4 in the electronic supplement, http://www.atmos-chem-phys.net/10/3297/2010/acp-10-3297-2010-supplement.pdf). While the dipole feature of transport over the Pacific is similar in the two reanalysis fields, we find that both downwind from dust source regions (south of 45° N at 500 hPa) and over the northeastern China (north of 45° N at 500 hPa), the northerly wind is stronger in GEOS-4 than NCEP toward the Pacific High. Stronger northerly wind of GEOS-4 is also observed at 900 hPa near 45° N. Since the fast transport is at the center of the dipole structure, it is conceivable that a relatively small changes in the transport in the model could lead to drastic changes in the trans-Pacific transport pathway. The GEOS-Chem model currently cannot use the meteorological data from the NCEP reanalysis, so we cannot test the sensitivity. We applied instead the HYSPLIT (Draxler et al., 1997) to compute the forward trajectories from the two most active deserts, Gobi and Taklimakan deserts. Since the HYSPLIT model cannot use the NCEP reanalysis winds, we used the NCEP Final Analysis (FNL) meteorological fields. The calculated trajectories are in better agreement with MODIS AOD observations than our simulation of dust transport (Fig. S5 in the electronic supplement, http://www.atmos-chem-phys.net/10/3297/2010/acp-10-3297-2010-supplement.pdf).

Another possibility is that the dust source regions prescribed in the model have a bias. If dust sources extended further north or northeast, closer to Siberia, a significant biomass burning source region of CO, EC, and OC, transport of dust in the model would be similar to CO and other aerosols and in closer agreement with the satellite observations. This study used the GOCART source function with latest Chinese desert maps from the ACE-Asia study (Chin et al., 2003), which include the recent desertification areas in the eastern Inner Mongolia (east of 110° E). We did not find newer desertification maps in China to use in this study.

7 Conclusions

MOPITT and MODIS observations show three events of rapid trans-Pacific transport in May 2003. We applied the global GEOS-Chem model to simulate these events. We find significant model biases in the first of the three transport events. While the observed transport pathways of CO and aerosols are generally consistent, enhancements of CO over the tropical Pacific are much broader than MODIS AOD enhancements. On the basis of model sensitivity studies, we find that the major fraction of the subtropical CO enhancements in the first half of May is due to biomass burning in Southeast Asia in April. Surface observations and model simulations of CO at the GMI site from the NOAA ESRL network provide corroborating evidence. Biomass burning CO was recirculated into the subtropical high-pressure systems and lingered for a much longer period than aerosols transported at higher latitudes. The biomass burning emissions from the Southeast Asia appear to be substantially underestimated in the GFED inventory in April 2003.

AOD enhancements are due to a combination of dust, sulfate, and organic and elemental carbons. Model results indicate that dust transport takes place at higher altitude than the other aerosols. Of the three transport events, simulated AOD transport has a clear bias in the first event. We attribute the bias to the transport of dust in the model. The model sensitivity is driven by a dipole structure over the Pacific with clockwise high-pressure system (the Pacific High) to the south and counter-clockwise low-pressure system (the Aleutian Low) to the north. The most rapid transport occurs in between the high-low pressure systems. As a result, relatively small changes of upwind source or transport could lead to a large change (as a result of bifurcation) in the transport pathway. CO and the other aerosols are transported mostly through the Aleutian Low (counter-clockwise system veering to the north), while dust in the model is transported through the Pacific High (clockwise system veering to the south). Stronger transport toward the Pacific High in GEOS-4 meteorological fields than in the NCEP reanalysis and in the FNL meteorological fields is likely a major factor contributing to the model bias.

Depending on the location of the dipole structure, trans-Pacific transport pathways simulated in the model could be very sensitive to model parameters such as up-stream wind field and source location, which may have large uncertainties. The resulting impacts on the simulated source-receptor relationship of pollutant transport from Asia to North America could be large. The dipole structure therefore poses a challenge on the modeling capability to simulate the transport pathways of pollutants across the Pacific and to project the impacts of Asian pollutants on North America.

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