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Seasonal variation of trans-Pacific transport of carbon monoxide (CO) in the upper troposphere: MLS observations and GEOS-Chem and GEM-AQ simulations

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Abstract

Multi-year Microwave Limb Sounder (MLS) carbon monoxide (CO) measurements at 215 hPa are employed to present a climatological view of seasonal variation of upper tropospheric trans-Pacific transport of Asian air pollution. The measurements show that the transport peaks in late boreal spring and early boreal summer. Although the strongest Asian air pollution outflow occurs in boreal summer, the "transport pathway" over the northeast Pacific is narrower in summer than in spring. Results from two tropospheric chemistry models GEOS-Chem and GEM-AQ are compared to MLS observations. Both models reproduce the strong trans-Pacific transport in boreal spring and summer well, but show different morphologies over Southeast Asia in winter and fall. A tagged CO simulation using GEOS-Chem indicates that Asian fossil fuel is the biggest source of upper tropospheric CO over the north Pacific in all seasons, excepting methane (CH_4) and non-methane hydrocarbons, although there are large fires in Southeast Asia in boreal spring and fall. A sensitivity test indicates that deep convection has a large effect on upper tropospheric CO abundances, increasing the abundances by more than 40%, over the north Pacific in boreal spring. In boreal summer, however, the increase is not significant over the north Pacific although it is large over continental Asia.

1 Introduction

Carbon monoxide (CO) is an important tropospheric trace gas. It is produced from fossil fuel, biomass burning, and the oxidation of methane (CH_4) and non-methane hydrocarbons (NMHC) (e.g., Duncan et al, 2007). It is mainly removed via reaction with hydroxyl radical (OH). Therefore, topospheric CO abundances is much higher in the Northern Hemisphere (NH) than in the Southern Hemisphere (SH) because of larger anthropogenic emissons in the NH than in the SH, and is higher in winter than summer due to seasonal variaitons of OH abundances (e.g., Novelli et al., 2003; Edwards et

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al., 2004). OH controls tropospheric oxidation capacity, and CO, whose reaction with OH is the main sink of that radical, affects the capacity. CO abundance also affects tropospheric ozone production, depending on the abundance of nitrogen oxides (NOx) (e.g., Logan et al., 1981; Crutzen et al., 1985). In addition, it is commonly used as a tracer of pollution transport because of its long chemical lifetime, about a couple of months in the troposphere (Folkins et al., 2006).

Trans-Pacific transport of air pollution has drawn extensive attention over the past two decades because of the fast economic growth in South and East Asia (e.g., National Research Council, 2009, and references therein). Most previous studies, mainly using aircraft measurements and/or model simulations, have focused on the NH spring, when there is strong transport of air pollutants in the lower and middle troposphere (e.g., Jacob et al., 2003; Bertschi et al., 2004; Singh et al., 2009; Zhang et al., 2008; Walker et al., 2010). Tropospheric background ozone abundances have increased since the 1970s over North America in the NH spring because of the trans-Pacific transport of Asian air pollution (Cooper et al., 2010). Jaffe et al. (2004) and Bertschi and Jaffe (2005), report long-range transport of Asian (Siberian) biomass burning air pollutants to western North America in the NH summer, and demonstrate that this could contribute to an exceedance of the ozone air quality standard in this region. Liang et al. (2007) also reported trans-Pacific transport events of Asian air pollution in the NH summer.

Using model results, Liang et al. (2004) show multiple transport pathways in the lower, middle, and upper troposphere, and suggest there is no evident seasonal difference in the frequency of transport events in the middle and upper troposphere although the magnitude of Asian CO abundances is larger in boreal spring than in boreal summer. Using airborne measurements over north America during February–May 2000, however, Wang et al. (2006) find that there is an enhancement of transport in the upper troposphere (UT) in late spring, not captured in their models. Jiang et al. (2007), using satellite observations between years 2004–2006, show strong upper tropospheric CO enhancement in the NH summer compared to in the NH spring, and link that with lofting of Asian pollutants into the UT by deep convection (e.g., Li et al., 2005; Fu et

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al., 2006; Park et al., 2009). However, the effect of deep convection on upper tropospheric trans-Pacific transport and the contribution of Asian emissions have not been quantified.

In this study, we use both satellite observations and model simulations to further study trans-Pacific transport in the UT. Aura/Microwave Limb Sounder (MLS) CO observations (Livesey et al., 2008) made between August 2004 and August 2010 are used to provide a climatological view of trans-Pacific transport of air pollution. Simulations made by two state-of-the-art global chemistry models the GEOS-Chem (Bey et al., 2001b) and the Global Environmental Multiscale Air Quality model (GEM-AQ) (Kaminski et al., 2008) are then compared with MLS CO observations over the north Pacific in the UT. Finally, we use GEOS-Chem to analyze the source regions and source types contributing to upper tropospheric CO, and to investigate the effect of deep convection on trans-Pacific transport of Asian air pollution.

The paper is presented in the following manner: A short description of MLS CO data, and the GEOS-Chem and GEM-AQ models is provided in Sect. 2. The seasonal variations in MLS observations and GEOS-Chem and GEM-AQ simulations in the UT are presented in Sect. 3. Upper tropospheric CO sources and the effects of deep convection on transpacific transport are studied in Sect. 4 and Sect. 5, respectively. A summary is provided in Sect. 6.

2 Measurements and model description

The MLS instrument (Waters et al., 2004) on the Aura satellite, launched in July 2004, focuses on upper tropospheric and middle atmospheric chemistry and climate, and measures profiles of temperature, ozone (O_3), CO, and about a dozen other chemical species from the UT to the mesosphere as well as ice water content (IWC) in the UT (Livesey et al., 2006). CO is retrieved from radiance measurements in the 240 GHz spectra region. MLS CO measurements have given rise to several important discoveries such as the trapping of pollution in the UT over South Asia (Li et

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al., 2005; Fu et al., 2006; Park et al., 2009), the CO tape recorder in the lower tropical stratosphere (Schoeberl et al., 2006), and the connections among surface emissions, convective uplifting, and long-range transport of CO in the UT (Jiang et al., 2007). Validation studies show that the MLS version 2.2 CO measurements at 215 hPa used here have a factor of ∼2 positive bias and a noise on individual profiles of 10–20 ppbv (parts per billion by volume) (Livesey et al., 2008). However, that study also shows that these CO measurements represent the CO morphology well at this level. In order to approximately correct the bias, all MLS CO volume mixing ratios (vmrs) at 215 hPa are divided by two in this study. In addition, we focus on the morphologies of CO distribution instead of absolute values when comparing model results to MLS observation in this study.

GEOS-Chem is a widely used chemistry transport model (CTM) with detailed tropospheric gas-phase and aerosol chemistry (Bey et al., 2001b). It is driven by assimilated Goddard Earth Observing System (GEOS) meteorology. With the fifth version of GEOS (GEOS-5) (Rienecker et al., 2007) meteorological data used in this study, GEOS-Chem has 47 hybrid sigma-pressure levels from the surface to 0.01 hPa (about 80 km). There are 34 vertical levels below 100 hPa (about 16 km) and the vertical resolution is about 1 km in the UT and lower stratosphere (LS).

In this study, GEOS-Chem version 8-03-02 is employed to conduct a full chemistry simulation for the period January 2005–December 2006 with a one year spin-up. The simulation is conducted on a $2^{\circ}\times2.5^{\circ}$ latitude by longitude grid. LINOZ parametrization of stratospheric ozone (McLinden et al., 2000) and dynamical tropopause are implemented in the model (D. Jones, personal communication, 2010). Deep convection is parametrized according to the Relaxed Arakawa-Schubert scheme (Moorthi and Suarez, 1992). CO sources includes anthropogenic, biomass burning, biofuel, and biogenic emissions and the oxidation from methane (CH_4). Liu et al. (2010) summarized recent updates of the CO anthropogenic and biomass burning emissions in the model. We note that in our simulations the biomass burning emissions are year-specific and are updated every 8 days from the version 2 Global Fire Emission Database

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(Randerson et al., 2007). In order to investigate contributions of various CO sources over Asia and other regions, a separate tagged-CO modeling technique (Duncan et al., 2007) implemented in GEOS-Chem is also employed in this study, as described in Sect. 4.

GEOS-Chem has been extensively used in studies of trans-Pacific transport of air pollution along with aircraft and satellite measurements (e.g., Bey et al., 2001a; Liu et al., 2003; Heald et al., 2003, 2006; Liang et al., 2004, 2007; Zhang et al., 2008). However, most of these studies focus on transport events in the lower and free troposphere. A climatological comparison of model results with observations in the upper troposphere has not been conducted. Liu et al. (2010) show deep convection is weaker and delayed in GEOS-5 in tropical regions, compared with GEOS-4 in which the Zhang-McFarlane deep convection scheme (Zhang and McFarlane, 1995) is used. In our study, we investigate seasonal variations in the effect of deep convection on trans-Pacific transport using GEOS-5 meteorology. Detailed evaluation of different deep convection schemes is beyond the scope of this study.

GEM-AQ is a global tropospheric chemistry with general circulation model (Kaminski et al., 2008) based on the three-dimensional Global Environemtal Multiscael model (GEM) developed by the Meteorological Service of Canada for operational weather forecasting (Côté et al., 1998). GEM-AQ has been used in various studies such as the Québec forest fires of July 2002 (O'Neill et al., 2006), air quality over Europe during the 2006 heat wave (Struzewska and Kaminski, 2008), the global distribution of hydrogen cyanide (HCN) in the UT (Lupu at al., 2009), inter-continental source–receptor relationships for ozone pollution (Fiore et al., 2009), and Arctic boundary-layer bromine chemistry (Toyota et al., 2010). In this study, the ability of the model to simulate upper tropospheric trans-Pacific transport of Asian pollution is evaluated against MLS observations.

The current simulation was conducted for the period January 2004–December 2006 on a $1.5^{\circ} \times 1.5^{\circ}$ global grid with 28 hybrid sigma-pressure vertical levels from the surface to 10 hPa (about 32 km). The vertical resolution is about 1 km in the UT/LS. Assimilated

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meteorology was used to update the meteorological fields every 24 hours. The Zhang-McFarlane deep convection scheme (Zhang and McFarlane, 1995) was used in this run. Unlike GEOS-Chem, which has higher anthropogenic emissions in the NH winter than in other seasons (Kopacz et al., 2010), the GEM-AQ anthropogenic emissions are constant throughout the year. Biomass burning emissions are the same as in the GEOS-Chem simulations.

3 Seasonal variations of MLS, GEOS-Chem, and GEM-AQ CO in the upper troposphere

3.1 Seasonal variation of transpacific transport as seen by MLS

Figure 1 shows latitude-time cross-section of MLS CO measured over the west Pacific (145°–155° E) and the east Pacific (215°–225° E, or 145°–135° W) at 215 hPa between 1 January and 31 December. The MLS measurements are averaged for the period August 2004–August 2010.

Over the west Pacific, MLS observations show the largest upper tropospheric CO around 30° N in boreal summer (June-July-August, JJA). During the period of January–March, relatively high CO abundances are seen in the tropics. This is related to the strong biomass burning over Southeast Asia in spring (van der Werf et al., 2003) and fast upward transport of pollutants produced in the lower troposphere in the tropics. CO abundances significantly increase in the northern subtropics during boreal spring (March-April-May, MAM) and maximize in both the northern subtropics and northern extra-tropics in boreal summer. This is thought to be caused by deep convection which lofts Southeast Asia biomass burning pollutants and continental Asian anthropogenic pollutants into the UT, and subsequent eastward transport over the northwest Pacific in the Asian monsoon season (Jiang et al., 2007; Liu et al., 2003). CO abundances in the northern subtropics and extra-tropics decrease in NH fall (September-October-November, SON), suggesting a decrease of the Asian outflow to the UT from

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weakening of deep convection over the Asian continent. The high CO near the Equator in October–November is due to biomass burning over Southeast Asia. We note, the seaonal and meridional variations of CO abundances in the UT differ significantely from these in the lower troposphere. In the lower troposphere, CO abundances increase with latitudes and the maximum CO abundances occurs at the northern high latiudes NH winter (Novelli et al., 2003; Edwards et al., 2004).

Over the east Pacific, large upper tropospheric CO abundances occur in subtropics and extra-tropics in spring and summer. CO abundances have little temporal change before mid-March, after which large CO abundances are seen. Considering the enhancement of Asian air pollution outflow over the west Pacific in the NH spring, the large CO abundances over the northeast Pacific demonstrate strengthened trans-Pacific transport in boreal spring compared to winter. CO abundances are large over a broad latitude range from the Equator to ~40° N in boreal spring, although the highest CO vmrs are centered at around 30° N in that season. The largest CO abundances occur in May and June, after which the large CO vmr "belt" moves northward and locates between 30° N and 50° N in July–August. This shifting of the high CO belt from spring to summer is driven by the northward movement of the subtropical westerly jet (not shown). CO abundances over the northern subtropics and mid-latitudes decreases in September and remain relatively low until the end of the year.

This study differs from the analysis in Jiang et al. (2007) which find that the strongest trans-Pacific transport of Asian pollution in boreal summer. We show the strongest trans-Pacific transport, as reported by the largest vmrs over the east Pacific, actually occurs in late boreal spring and early summer (May–June). The discrepancy is due to different analysis methods. In Jiang et al. (2007), the seasonal variation is derived from monthly means over the northeast Pacific region between 20°–45° N at 150 hPa where MLS most likely measures the lower stratosphere in NH winter and measures the upper troposphere in NH summer, therefore the seasonal variations of CO abundances is affected by the seasonal variations of the tropopause. Wang et al. (2006) show enhanced trans-Pacific transport in the UT in late spring from airborne measurements,

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but these measurements were not made after May 2000. In addition, we show air with large CO abundances reaches North America in the northern tropics and subtropics in spring but only in the extra-tropics during summer, which is consistent with the finding that back trajectories of measured Asian air pollution in the UT over North America always pass over the northwestern United States in summer (Liang et al., 2007).

3.2 Comparison of GEOS-Chem and GEM-AQ simulations to MLS observations

In order to evaluate the GEOS-Chem and GEM-AQ representations of trans-Pacific transport of air pollution in the UT, Fig. 2 shows two-year (2005–2006) seasonal means of MLS measurements at 215 hPa, and of model results sampled at locations of MLS observations and interpolated at 215 hPa over Asia, the north Pacific, and North America. Fig. 2 also shows two-year seasonal mean horizontal winds from National Centers for Environmental Prediction (NCEP, Kalney et al., 1996), GEOS-5, and GEM, as well as MLS IWC, used as an index of deep convection (Jiang et al., 2007). We note that MLS averaging kernels are not applied to the model results here. Because MLS averaging kernels are generally narrow, the difference between model results with and without application of the average kernels is very small, and it is generally less than 4% at lower and mid-latitudes. In addition, comparison shows that seasonal means for these two years are representative of seasonal means for the full six-year record with differences generally less than 4% except over Southeast Asia where the difference is as large as 12% in SON. The large difference over Southeast Asia is due to the unusual strong fire over Indonesia in October–November 2006 (Logan et al., 2008; Nassar et al., 2009). The seasonal standard deviations of CO observations and simulations are shown in Fig. 3. Because MLS CO at 215 hPa has a noise of ~20 ppbv (in measurements without the approximate factor of two bias correction), normally distributed noises with a mean value of 10 ppbv are added to GEOS-Chem and GEM-AQ results at 215 hPa before the standard deviations are computed in order to aid the Fig. 3 comparisons.

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GEOS-Chem and GEM-AQ seasonal means show good agreement with MLS morphology over the north Pacific in the NH winter, when trans-Pacific transport is weak. GEOS-Chem results show a similar morphology of the large CO vmrs centered over Southeast Asia as MLS measurements. However, the absolute values for GEOS-Chem CO abundances are much larger than bias corrected MLS observations. GEOS-Chem results also have large standard deviation over Southeast Asia in winter (Fig. 3). Further analysis reveals that GEOS-Chem produces large CO in the middle and upper troposphere (not shown) over Southeast Asia in February–March 2005. GEM-AQ results, however, show high CO vmrs over the west India Ocean and India instead of over maritime Southeast Asia, indicating the vertical transport in this model needs to be improved. Despite these difference over South and Southeast Asia, both models produce slightly enhanced CO between 10° N and 30° N over the Pacific, demonstrating trans-Pacific transport at lower latitudes, as also shown by MLS observations in winter. The standard deviations (Fig. 3) are generally small in both observations and simulations over the Pacific in winter, suggesting that this transport is somewhat continuous rather than episodic.

In spring, both GEOS-Chem and GEM-AQ reproduce the strong trans-Pacific transport in the UT seen by MLS. GEOS-Chem produces a very similar transport pattern to MLS observations. Air with large CO vmrs is seen over Southeast Asia, south China, and the north Pacific south of 40° N in MLS observations and GEOS-Chem and GEM-AQ results. Because the southeast-ward movement of a branch of the subtropical westerly jet over the mid-Pacific, some air pollution is transported into lower latitudes and then is transported across Mexico. Airborne measurements also show this feature (Heald et al., 2003). In contrast to MLS observations and GEOS-Chem results, GEM-AQ results do not show high CO abundances over maritime Southeast Asia. The standard deviations in MLS, GEOS-Chem, and GEM-AQ results are slightly larger over the north Pacific in spring than in winter, and there is generally good agreement between the standard deviations of the measurements and of GEOS-Chem results. The extremely large standard deviations over maritime Southeast Asia in GEOS-Chem is

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due to strong episodic vertical transport of biomass burning emission in early 2005.

In summer, both GEOS-Chem and GEM-AQ simulations show the largest CO abundances over South China, as seen in MLS observations. GEM-AQ produces larger outflow of Asian CO over the northwest Pacific in summer than in spring, consistent with MLS observations. GEOS-Chem output, however, does not show a clear enhancement of CO over the west Pacific from spring to summer. Because of the upper tropospheric anticyclone over South and East Asia, MLS observations show evident westward transport over the Indian Ocean, which would result in high East Asian pollutant abundances over West Asia in the NH summer (Li et al., 2001; Stohl et al., 2003). At higher altitudes, air with high CO abundance is trapped in the center of the anticyclone over Tibetan Plateau and enventually is updrafted into the LS (Li et al., 2005; Fu et al., 2006; Park et al. 2009). The westward transport seen by MLS is only seen at the northern subtropics, not over the Indian Ocean, in either GEOS-Chem or GEM-AQ simulations. Nevertheless, both models show a narrow high CO plume between 30° N and 50° N over the northeast Pacific. The standard deviations (Fig. 3), in GEOS-Chem results are comparable to MLS observations and they are larger in GEM-AQ results than in MLS observations.

In fall, GEOS-Chem results are in generally good agreement with MLS observations over Southeast Asia and the north Pacific; however, GEM-AQ results significantly differ from MLS observations. MLS measurements show large mean CO vmrs and standard deviations over maritime Southeast Asia, reflecting the large fires over Indonesia in October–November 2006 (Logan et al., 2008; Nassar et al., 2009). GEOS-Chem CO abundances and standard deviations are larger than MLS CO abundances over maritime Southeast Asia and over Bangladesh. However, both MLS and GEOS-Chem show weakened trans-Pacific transport of CO during fall compared to during summer. GEM-AQ CO abundances are significantly smaller than MLS CO abundances over maritime Southeast Asia, but much larger than MLS observations over Bangladesh. In addition, GEM-AQ has larger standard deviations than MLS over the Indian Ocean and South Asia. The disagreements between the locations of the maximum GEM-AQ and

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MLS CO abundances and standard deviations suggest that convective parameterization, which is need to loft surface biomass emissions to the UT in models (Taguchi et al., 2002; Duncan et al., 2003), in GEM-AQ needs improvements.

Recent studies show that chemistry transport model results in the upper troposphere are sensitive to the deep convection schemes as different deep convection parametrization schemes produces different upward mass fluxes (Barret et al., 2010; Williams et al., 2010). Liu et al. (2010) show that GEOS-Chem simulations with the Zhang-McFarlane deep convection scheme (Zhang and McFarlane, 1995) are in better agreement with space-borne observations than simulations using Relaxed Arakawa-Schubert scheme (Moorthi and Suarez, 1992) in tropics. Further studies using chemical measurements in the upper troposphere are needed and should produce more insights into the parametrization of deep convection used in weather forecast and climate ressearch models (Folkins, et al., 2006).

4 Upper tropospheric CO source analysis

A GEOS-Chem tagged CO simulation (Duncan et al., 2007) has been conducted to identify the origins of upper tropospheric CO. In this simulation, anthropogenic and biomass burning CO emissions in various regions as well as the products from the oxidation of CH_4 and a few NMHC are tagged as different CO species. Archived monthly mean OH is used to calculate CO production and loss rates. This type of simulation has been conducted in many studies such as CO budget estimates using ground data (Duncan et al., 2007) and inversion (top-down) estimation using aircraft and satellite observations (Palmer et al., 2003; Heald et al., 2004; Arellano et al., 2006; Kopacz et al., 2010).

In the current study, a simulation for 2005–2006 is analyzed. Seasonal means of total CO, fossil fuel CO emissions in Asia (70° E–152.5° E, 8° N–45° N, see Fig. 4 for the geography of source regions) and in the rest of the world, biomass burning CO emissions in Asia and Oceania (including Australia, 70° E–170° E, 90° S–45° N), in Africa

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(17.5° W–70° E, 48° S–36° N), and in the other regions in the world, and CO produced from the oxidation of CH_4 and NMHC, as well as CO made by MLS for the same period are shown in Fig. 5. Although simulated total CO shows stronger seasonal variations than MLS observations in winter-spring-summer over maritime Southeast Asia, model results generally show good agreement with MLS observations in term of CO abundances and seasonal variations over South and East Asia, the northwest Pacific, and the northeast Pacific. This suggest that the tagged CO simulation is capable of reproducing upper tropospheric CO even though the interaction of OH and CO, especially important in the case of large fires (e.g., Duncan et al., 2003), is ignored.

It can be seen from Fig. 5 that CH_4 is generally the dominant source of upper tropospheric CO except over South and East Asia in summer. The CH_4 contribution to total CO is about 29–38% over all regions and seasons. Chemical production from CH_4 is slightly larger in boreal summer and fall than in winter and spring over South and East Asia (Panel B), the northwest Pacific (Panel C), and the northeast Pacific (Panel D). NMHC contributes ~20% of total CO in the UT, with small seasonal and regional variations. We note the regional sources of CH_4 and NMHC are not identified in this study. CH_4 is a long-lived species, CO produced from the oxidation of it can be either an in-situ product or have been transported for a long distance. NMHC are short-lived species, and little of them can survive a long-range transport. Therefore, the oxidations of NMHC most likely occur in the lower troposphere over continental Asia and maritime Southeast Asia.

Over maritime Southeast Asia (Fig. 5, Panel A), a prominent feature is the significant Asian and Oceanian biomass burning contribution to total CO in boreal fall. This is due to the large fires in Indonesia in fall 2006 (Logan et al., 2008, Nassar et al., 2009). Asian anthropogenic emission is the biggest source (excepting CH_4 and NMHC) in the other seasons. Climatologically, biomass burning emissions in this region are large in MAM (Van der Werf et al., 2003). As a result, the contribution of biomass burning emission is comparable to the contribution of Asian fossil fuel emissions in spring. Fossil fuel emissions in the rest of the world contribute about 6–10% in all seasons,

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and are almost the same as over South and East Asia, the northwest Pacific and the northeast Pacific. Biomass burning emissions in Africa and the other regions contribute a very small part, less than 7%, of the total CO in the UT.

Over South and East Asia, fossil fuel contribution to total CO is 17–19% in boreal winter and spring, and is significantly large in boreal summer (37%), and in boreal fall (27%). The increase from spring to summer is due to the enhancement in upward transport (Jiang et al, 2007) rather than increases of surface emissions. In fact, the anthropogenic emission is larger in winter than in summer (Kopacz et al., 2010). Asian and Oceanian biomass emissions are small contributors in all seasons, with a maximum of about 7% contribution in MAM. Biomass burning in Africa contributes 10% in DJF and 6% in MAM. These are sightly larger than and are comparable to the contributions of local biomass burning emissions in DJF and MAM, respectively, demonstrating the global transport of air pollution.

Over the northwest Pacific and the northeast Pacific, the relative contributions of various CO sources are close, although the absolute contributions are smaller over the northeast Pacific than over the northwest Pacific. Asian fossil fuel contributes 16–17% in winter, 20–22% in spring and fall, and 26–32% in summer over both the northwest and northeast Pacific. Asian and Oceania (including Australia) biomass burning contributions are small in winter (4–5%) and summer (2%). They are about 8–9% in spring which is comparable to the contribution of fossil fuel emissions in the rest of the world. In fall, they only contribute 7% over the northwest Pacific and 5% over the northeast Pacific even though they contribute 21% over maritime Southeast Asia. Biomass burning emissions in Africa contribute 8–9% in winter, 6% in spring and very small part in summer and fall. Biomass burning emission in the other regions contributes less than 5% in all seasons. In short, over the north Pacific, Asian fossil fuel is the largest source of upper tropospheric CO, excepting CH_4 and NMHC, in all seasons even though the most probable trans-Pacific transport pathway starts in Southeast Asia in boreal spring as discussed in Sect. 3.1. This differs from the discussion in Jiang et al. (2007) in which biomass burning emissions are thought to be the main component of CO transported

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across the north Pacific in MAM. However, this source analysis of upper tropospheric CO over the north is similar to a previous analysis that Asian anthropogenic CO emission is the largest contributor in Asian CO outflow mass flux in the free (mainly the lower and middle) troposphere, while anthropogenic emission in the other regions and African biomass burning emission only contribute small parts of the outflow, in all seasons (Liu et al., 2003).

5 Effects of deep convection

Deep convection plays an important role in lofting surface air pollutants into the UT (e.g., Folkins et al., 1997; Andreae et al., 2001; Taguchi et al., 2002; Li et al., 2005; Jiang et al., 2007) where horizontal wind speeds are larger and inter-hemisphere transport of air pollution is faster than in the lower troposphere. In particular, Jiang et al. (2007) linked strong trans-Pacific transport of Asian pollution in the UT with deep convection over Asia during the summer monsoon. In order to quantify its effect on trans-Pacific transport, a GEOS-Chem simulation has been conducted with the deep convection turned off. We note, however, that the driving meteorological data themselves contain the effects of deep convection. For example, convective influences on cloud and moisture are parametrized (e.g., Moorthi and Suarez, 1992). However, the deep convection is not resolved in the gridded wind data. In a chemical transport model (CTM), therefore, a deep convection parameterization is necessary to calculate subgrid convective mass flux besides wet scavenging of aerosols in clouds, and lightning production of NO_x, etc. (e.g., Liu et al., 2001; Williams et al., 2010). Recent studies show that simulation results from CTMs are sensitive to the deep convection schemes (Barret et al., 2010; Williams et al., 2010). As with those studies, we can assess the effect of deep convection using a CTM with the deep convection scheme turned off.

Seasonal means of CO simulated by GEOS-Chem with the deep convection turned off are shown in Fig. 6. High CO abundances are centered over the east tropical Indian Ocean in winter and spring, over India, south China, and the northwest Pacific in

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summer, and over South and Southeast Asia and tropical Indian Ocean in fall. However, the CO abundances over continental South and East Asia and maritime Southeast Asia are evidently smaller than in the simulation with the deep convective transport parametrization turned on (Fig. 2), especially in MAM and JJA. The large CO vmrs over tropical Indian Ocean in fall are similar to those obtained when the deep convection is turned on. This is due to upward transport of large CO emission during the large fires of 2006 (Logan et al., 2008; Nassar et al., 2009), but the extremely large CO vmrs (shown in Fig. 2) are missed over Sumatra.

The relative differences between the simulations with and without deep convection are also shown in Fig. 6. Deep convection significantly increases upper tropospheric CO abundances over continental South and East Asia continent and maritime Southeast Asia in all seasons. Over the north Pacific, however, it only significantly increases CO in DJF and MAM. In DJF, the maximum increase is above 50% over Sumatra, and above 30% over South and East Asia and the north Pacific. In spring, the enhancement caused by deep convection is above 50% over South and East Asia, and above 40 % over the north Pacific. The maximum enhancement by deep convection is smaller over continental South and East Asia (40–50%) and over the north Pacific (10–20%) in JJA than in MAM. Calculations (not shown) show significatly large positive CO convective mass flux over continental South and East Asia, but negtive CO convective mass flux over the Northwest Pacific in JJA, which explains why there is a large ehancement over continental South and East Asia but only a small ehancement over the nothwest Pacific in the NH summer. The negative CO convective mass flux is due to the low CO abundances in the lower troposphere and high CO abundances in the UT over the ocean. That is, deep convection over the northwest Pacific generally lofts clean air from the lower troposphere to the UT. Nevertheless, the simulation with deep convection turned off does produce higher CO abundances in JJA than in MAM over Asia and the Northwest Pacific. We expect strong upward large scale circulation within the intertropical convergence zone (ITCZ) (Waliser and Gautier, 1993), which sits over the Asian pollution source region in summer, plays an important role. In SON, deep

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convection has an even smaller effect on upper tropospheric CO than in JJA over the north Pacific. However, it increases CO by more than 80% over Indonesia where large fires occurred in October–November 2006. This confirms the important role that deep convection plays in producing the extremely high levels of CO in the UT, even though deep convection is rare in a drought period favoring large fires (Folkins et al., 1997; Andreae et al., 2001; Taguchi et al., 2002; Liu et al., 2010)

6 Summary

In this study, multi-year MLS CO measurements at 215 hPa are used to study the seasonal variation of trans-Pacific transport of Asian pollution in the UT. GOES-Chem and GEM-AQ outputs are compared to these observations. Contributions of different CO emission regions and types and the effects of deep convection are quantified using GEOS-Chem simulations.

Six years of MLS observations show strong trans-Pacific transport of air pollution in both boreal spring and summer, peaking in late spring and early summer. Asian pollution reaches to North America over a broad latitude range of 0°–40° N in spring. Although the strongest Asian outflow of air pollution occurs in summer (Jiang et al., 2007), it only reaches to North America at 30° N–50° N in that season.

The comparison between simulated and measured seasonal means shows that both models are in good agreement with observations, and are able to reproduce the strong trans-Pacific transport in the NH spring and summer. However, GEOS-Chem produces large CO vmrs over Southeast Asia in boreal winter and GEM-AQ produce large CO vmrs over continental South and East Asia in fall, in contrast to the morphology of MLS observations. The comparison shows that the deep convection parametrization in GEM-AQ needs to be improved. These large simulated CO abundances are related to the vertical transport of large fire emissions in early 2005 and fall 2006 in Southeast Asia. Detailed event studies are planned aiming to investigate the strong vertical transport in these simulations.

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A GEOS-Chem tagged CO simulation shows that (except for chemical production from CH_4 and NMHC) Asian fossil fuel is the biggest source of upper tropospheric CO over the north Pacific in all seasons. Its contribution to upper tropospheric CO over the north Pacific ranges from 16–17% in the NH winter to 27–30% in the NH summer. Asian and Oceanian biomass burning emissions contribute a small part, less than 10%, to the total CO in the UT even though it contribute more than 20% over southeast Asia in the NH fall.

A sensitivity test using GEOS-Chem, driven by GEOS-5 meteorology, reveals that deep convection plays an important role in lofting surface air pollutants into the UT in the NH spring. Over the north Pacific, the maximum convective enhancements are above 30% in the NH winter and above 40% in the NH spring. Although deep convection increases the upper tropospheric CO abundances by more than 40% over continental South and East Asia, it only slightly increases CO abundance transported over the north Pacific in the NH summer.

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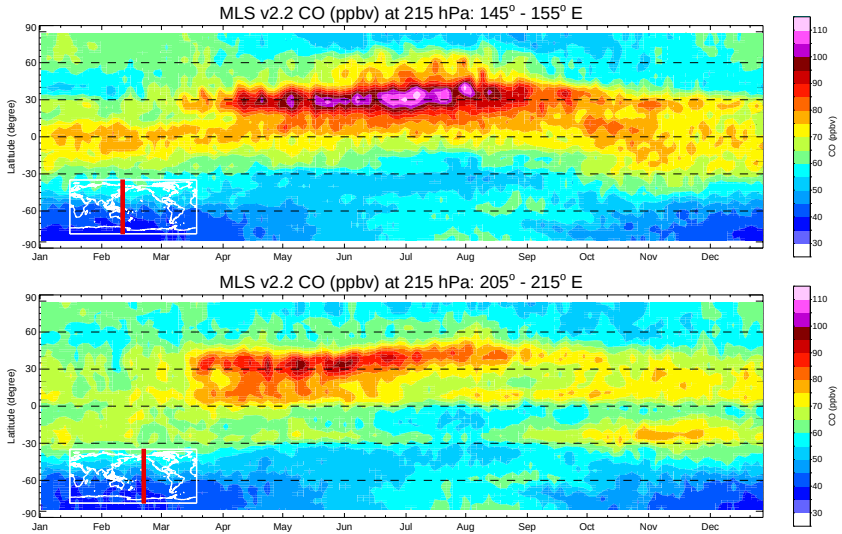

Fig. 1. Time-latitude cross-sections over (top panel) the west Pacific (145°–155° E) and (bottom panel) the east Pacific (215°–225° E) of MLS v2.2 CO averaged between August 2004–August 2010 at 215 hPa. MLS v2.2 CO vmrs are divided by two in order to approximately correct the bias at 215 hPa (As is also the case for all the following figures). The red squares in the inserted maps represents the locations of the slides shown.

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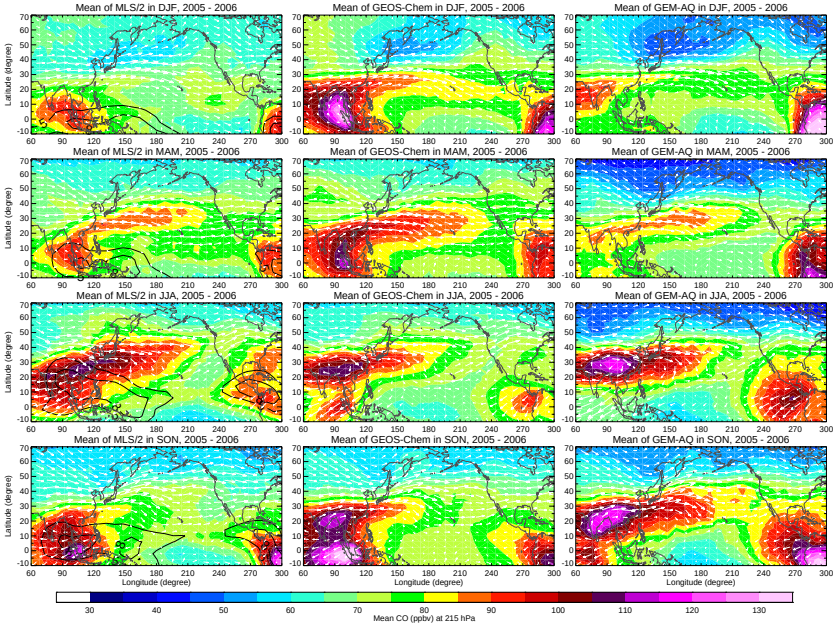

Fig. 2. Seasonal CO means from (left column) MLS v2.2 (devided by two), (middle column) GEOS-Chem, and (right column) GEM-AQ at 215 hPa in the Northern Hemisphere winter (DJF), spring (MAM), summer (JJA), and fall (SON). White vectors represent (left column) NCEP, (middle column) GEOS-5, and (right column) GEM horizontal winds. Black contours in the left column represent 5 mg/m^{-3} and 8 mg/m^{-3} MLS ice water content (IWC), index of deep convection.

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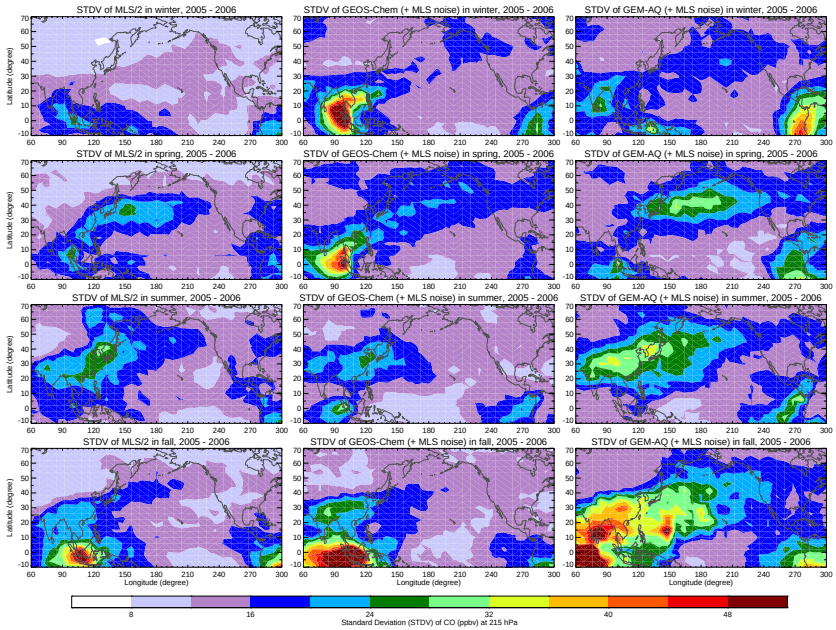

Fig. 3. Standard deviations of (left column) MLS v2.2 (divided by two), (middle column) GEOS-Chem, and (right column) GEM-AQ CO vmrs at 215 hPa. Gaussian noise with a 10 ppbv standard deviation is added to the model results for easy comparison to the MLS observations.

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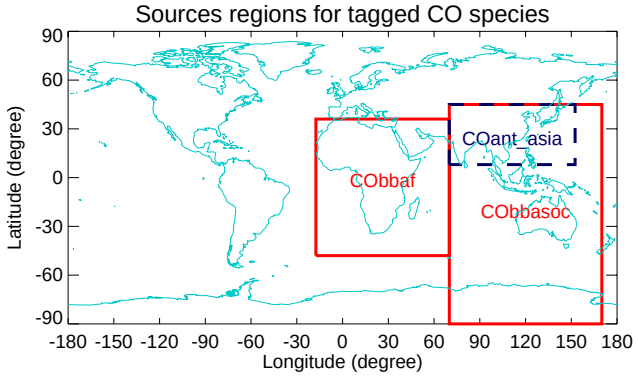

Fig. 4. Geography of source regions for some tagged CO species: COant_asia, fossil fuel CO emission in Asia (70° E–152.5° E, 8° N–45° N); CObbasoc, biomass burning CO emission in Asia and Oceania (70° E–170° E, 90° S–45° N); and CObbaf, biomass burning CO emission in Africa (17.5° W–70° E, 48° S–36° N).

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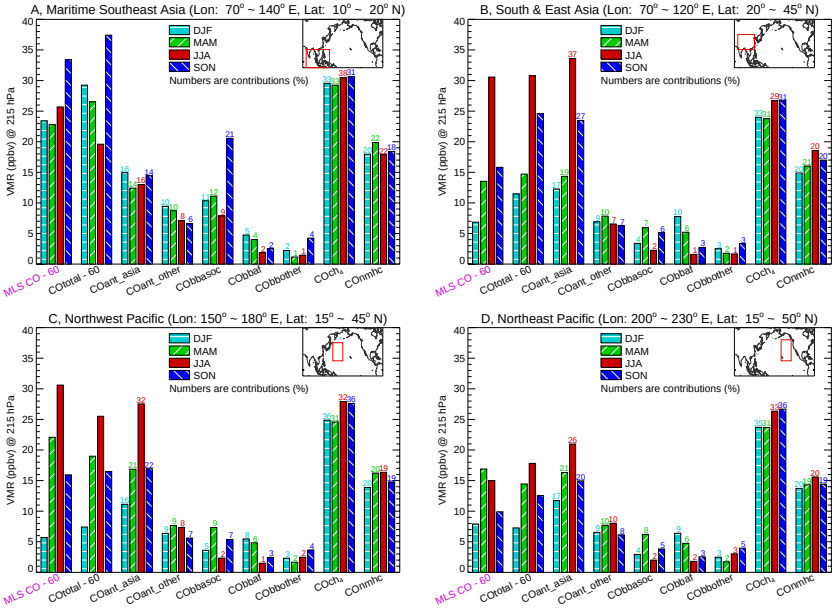

Fig. 5. Regional mean 215 hPa CO abundances from MLS v2.2 (divided by two) and from GEOS-Chem tagged CO simulations above **(A)** Maritime Southeast Asia; **(B)**, South and East Asia; **(C)** the northwest Pacific; and **(D)** the northeast Pacific. 60 ppbv has been subtracted from MLS CO and simulated total CO (COtotal) in order to fit the range of the y axis. *COant_asia* is fossil fuel CO emission in Asia; *COant_other* is fossil fuel CO emission in the other regions in the world. *CObbasoc* is biomass burning CO emission in Asia and Oceania; *CObbaf* is biomass burning CO emission in Africa; *CObbother* is biomass burning CO emission in the rest of the world; and $COch_4$ and *COnmhc* are CO production from CH_4 and non-methane hydrocarbons (NMHC), respectively. Geography of source regions for *COant_asia*, *CObbasoc* and *CObbaf* are shown in Fig. 4.

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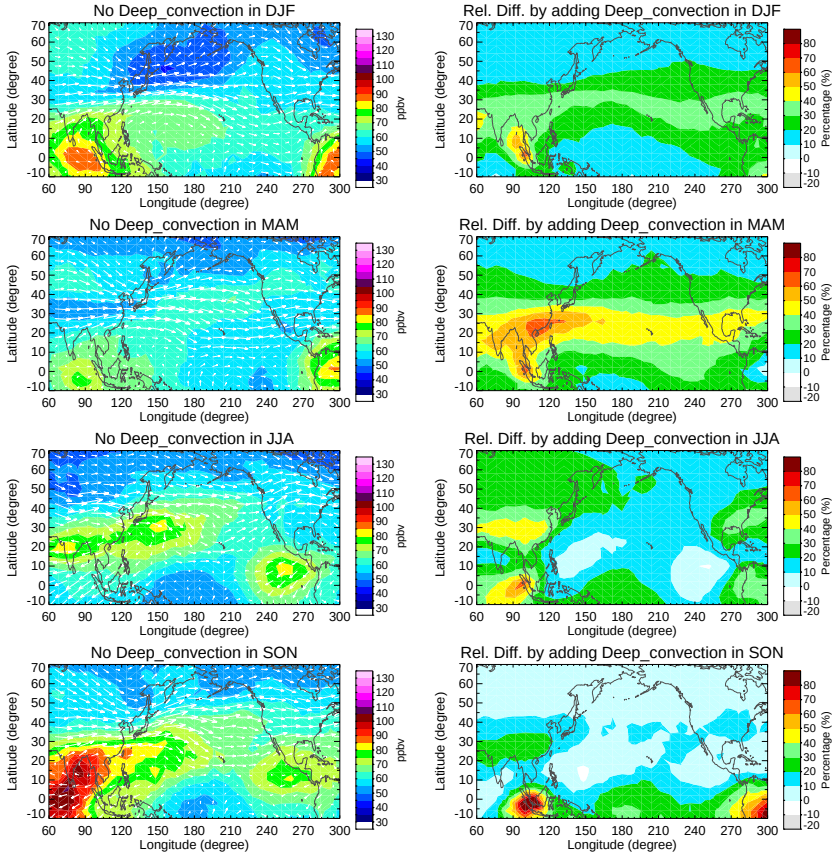

Fig. 6. (Left column) Seasonal means of GEOS-Chem CO abundance (ppbv) at 215 hPa and sampled at MLS measurement locations for years 2005–2006. Deep convection is turned off in this simulation. White vectors are NCEP horizontal wind at 200 hPa for the same period. (Right Column) Relative difference (%) ("With deep convection" minus "No deep convection" devided by "No deep convection") caused by adding deep convection.

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