Recent Springtime Regional CO Variability in Southern China and the Adjacent Ocean: Anthropogenic and Biomass Burning Contribution

Ka-Ming Wai1*, Peter A. Tanner2

1 Department of Geological and Mining Engineering and Sciences, Michigan Technological University, Houghton, MI, USA
2 Department of Science and Environmental Studies, The Hong Kong Institute of Education, 10 Lo Ping Road, Tai Po, New Territories, Hong Kong S.A.R., China

ABSTRACT

Springtime regional CO variability in the boundary layer and lower free troposphere in recent years has been studied by a combination of chemical transport models, satellite retrievals and surface-based measurements. Based upon the surface-level measurements and satellite observations, the CO mixing ratios and column densities downwind of the continental outflow were found to have significant reductions in the springtime of 2009 and 2010. Under the influence of continental outflow, there were 58% and 38% decreases in the monthly mean mixing ratios at a rural coastal site in March and April 2009–10, respectively, compared to the 2007–08 figure. Other than the global economic recession, the CO reductions downwind are attributed to the recent implementation of effective anthropogenic CO emissions control in China. The CO simulations by the MM5-CMAQ modeling system with fixed annual emissions support this argument. Features of frontal passage, including the higher CO mixing ratios in the post-frontal stage, were well-captured by the modeling system. The high-resolution nested-grid GEOS-Chem model was used to better characterize the downwind influences of CO due to the biomass burning in Southeast Asia in March and April 2008. The computed CO mixing ratios agreed well with satellite observations at 700 hPa during selected episodes when intense biomass burning activities in Southeast Asia were observed by the satellite images. The biomass-burning-derived CO provided a large contribution (33–86%) to the total CO mixing ratio in the lower free troposphere downwind over the South China Sea and western Pacific Ocean, based on the modeling results. Vertical export of CO in active convection events near the source region was evident.

Keywords: Continental outflow; China; Emissions control; Biomass burning; Chemical transport model.

INTRODUCTION

Carbon monoxide (CO) is an important tropospheric species which affects the oxidative capacity of the troposphere through its reaction with hydroxyl radicals. It is also a toxic gas and affects human health when its mixing ratios are high (> 6 ppm) at the surface level. Emissions from combustion of fossil fuels (from vehicles and industrial activities) and biomass burning (bb)/wildfires are the major primary sources of CO (Jaffe et al., 1997; Andreae and Merlet, 2001; Edwards et al., 2004).

Heavy air pollution loadings associated with the recent rapid growth of the economy in China in the past few decades have been well-documented. Pollution outflow from continental East Asia to nearby remote areas has been studied using surface-level/aircraft measurements, chemical transport models and remote sensing techniques (Heald et al., 2003; Russo et al., 2003; Lin et al., 2011). During the TRACE-P campaign, backward trajectories were utilized to identify five principal Asian source regions of outflow. The maximum CO mixing ratio was more than a factor of 2 larger, compared to the background, in the boundary layer of the central and coastal regions due to the industrial activities in East Asia (Russo et al., 2003). Trans-Pacific transport of Asian CO pollution in spring 2001 was also studied. The O3 enhancements were found only in some Asian CO plumes (Heald et al., 2003). The continental outflow was also studied at an elevated remote site in Taiwan. The monthly means of both CO and O3 mixing ratios showed maxima in spring and in the continental air masses from Southeast Asia, coastal China, and Korea/Japan (Lin et al., 2011). However, along with the recent focus upon emissions control and technology improvement in China (He et al., 2010) a reduction in air pollution has been reported (Witte et al., 2009; Li et al., 2010; Zhou et al., 2010). Significantly, the daily urban traffic emissions of CO in Beijing were...
reduced by 44.5% compared with the levels before the 2008 Olympic Games, partly due to better traffic management during the Games (Zhou et al., 2010). During the same period, satellite measurements showed a CO reduction of 12% at 700 hPa, when Beijing and neighboring provinces to the south were included, compared to the past three years (Witte et al., 2009). For other pollutants, dramatic reductions of SO₂ emissions were observed by the space-borne Ozone Monitoring Instrument (OMI) in 2008 in several areas in northern China where large coal-fired power plants operate. These reductions confirmed the effectiveness of the control measures (Li et al., 2010). Whether or not such control measures and technology improvements have effects upon remote areas through the outflow of CO is not established, and thus an update is urgently needed.

Southeast Asia is one of the major sources of bb emissions in the world (Streets et al., 2004), with peak bb activities taking place in springtime (i.e., a similar period as above). The bb-derived CO dominates the total CO emissions within the region. The prevailing wind in this region is, in general, westerly for this time of the year, and the smoke is carried to the east (Hsu et al., 2003). The downwind impacts on southern China were reported according to the O₃ enhancement in sounding profiles (2.5–4.5 km above ground) from the results of a backward trajectory model (Liu et al., 1999). The bbs in Southeast Asia, which were particularly intense in spring 1999 during the PEM-Tropics B campaign, contributed most of the CO enhancements observed in the free troposphere over the northern tropical Pacific (Staudt et al., 2001). Based on aircraft samples collected over the Western Pacific Ocean during the TRACE-P campaign at latitudes below approximately 25°N, enhanced CO and fine-particle aerosol mixing ratios were observed in the plumes. Back trajectories and satellite fire map data suggested their bb origin from Southeast Asia. Purest biomass burning plumes were found in layers at altitudes from 2 to 4 km asl. (Ma et al., 2003). The impacts of bb were also detected at an elevated site (~2860 m) in Taiwan (Wai et al., 2008) during springtime. The change of particle size due to the bb plumes was studied with the AERONET dataset in southern China (Wai and Tanner, 2010). It is noted that boundary layer outflow over the western Pacific during springtime is largely devoid of bb influence (Liu et al., 2003), because of the dominant effects of the East Asian Monsoon at the boundary-layer levels (Wai and Tanner, 2010). It is noted that most of the previous studies focused on short time periods and very few events. Therefore, more studies are required to better characterize the downwind impacts of bb due to the inter-annual variability of bb emissions (Van der Werf et al., 2006) and meteorology (i.e., vertical export of pollution).

The objective of the present study is to investigate recent springtime regional CO variations in the boundary layer and lower free troposphere, with a combination of tools including chemical transport models, remote sensing products and surface-level measurements. The inter-annual CO variations in springtime 2007–2010 at downwind locations of the continental outflow in East Asia are firstly investigated. The possible explanation of the variations is discussed in detail. The regional impacts of bb-derived CO and its contribution to the total CO mixing ratio in the lower free troposphere in spring 2008 are then studied by a high-resolution model, based on the latest available bb emissions inventory. The fine structure of the bb plume is better resolved, compared with that produced by coarse-grid (horizontal resolution of 2°×2.5° or 4°×5°) models used in previous studies mentioned above.

METHODS

Observational Data

Surface-level CO mixing ratios were measured by a commercial instrument TECO 48C, API 300 with the working principle of non-dispersive infra-red absorption with gas filter correlation. The monitoring site is of rural nature and located in Hong Kong-Tap Mun (22.5°N, 114.3°E, Fig. 1), southern China and is free from the influence of local emissions. Carbon monoxide measurements at this site have been utilized elsewhere, such as in Wang et al. (2006). The site is operated by the Hong Kong Environmental Protection Department with quality control measures detailed in EPD (2011).

For the satellite retrievals, the CO distributions in the lower free troposphere were obtained from MOPITT (Measurements of Pollution in the Troposphere) Level-3 daily daytime products (1°×1° gridded) at 700 hPa. MOPITT has poor sensitivity in the boundary layer. The MOPITT retrievals have been used extensively in bb, regional and hemispheric pollution studies (Heald et al., 2003; Edwards et al., 2004; Edwards et al., 2006). The CO column densities from the AIRS (Atmospheric Infrared Sounder) global 1°×1° monthly Level-3 products were also used. Bb events in Southeast Asia were verified by MODIS (Moderate Resolution Imaging Spectro-radiometer) imagery for thermal anomaly and 10-days global fire map.

Description of Numerical Models

To deal with the two aspects of CO regional transport in Asia with the different source origins (i.e., China and Southeast Asia) mentioned in the previous section, two chemical transport models were adopted separately. The USEPA Community Multi-scale Air Quality (CMAQ v4.7; Byun and Ching, 1999) Model was adopted to quantitatively study the impacts of boundary-layer continental outflow of anthropogenic CO in East Asia on downwind southern China. The CMAQ model has been employed in many air quality studies including those conducted recently in East Asia and China (An et al., 2007; Chen et al., 2007; Liu et al., 2010a, b; Liu et al., 2012). The meteorological fields of the study periods were provided by the National Center for Atmospheric Research (NCAR)/Pennsylvania State University (PSU) Fifth-Generation Mesoscale Model (MM5 v3.7, Grell et al., 1994) and processed by the Meteorological-Chemical Interface Processor (MCIP v3.3) for CMAQ-ready inputs. Physical options of the MM5 model such as Grell’s cumulus option, MRF PBL, Simple Ice (Dudhia) explicit precipitation, RRTM long-wave
radiation schemes and 5-layer soil model were applied. The synoptic-scale temperature distribution, wind fields and location of mid-latitude cyclone during the study periods are comparable to those shown in the surface pressure maps and predictions by the NCEP Global Data Assimilation System (GDAS) model. The emissions were based on the NASA’s INTEX-B project emissions inventory (Zhang et al., 2009) in 2006, which included major anthropogenic pollutants such as SO₂, NOₓ, CO, and 30 lumped VOC species with 0.5° × 0.5° resolution. Biogenic emissions of non-methane volatile organic compounds (NMVOC) were calculated by the MEGAN model (Guenther et al., 2006). The CMAQ model domain covered East Asia including China, Korea, Japan and Southeast Asia. The horizontal grid resolution of 27 km × 27 km was configured with 26 vertical layers spanning from the surface to 50 mb, with less than 25 m each for the first 5 vertical layers. The boundary conditions were supplied by the outputs of the GEOS-Chem global chemistry model. A 6-day spin-up period was used in the study to minimize the effect of the initial conditions. Other model settings were detailed in Liu et al. (2012). With these settings, the regional SO₂ transport in northern China was successfully captured, and the results agreed with the observations made at an elevated site (Liu et al., 2012).

To better characterize the bb emissions in Southeast Asia and resolve the fine structure of the bb plume downwind, the high-resolution nested-grid GEOS-Chem model (0.5° × 0.667° or 57 km × 74 km) was adopted. The nested model has been used to study regional CO pollution and its export from China (Chen et al., 2009) and surface CO pollution in Europe (Protonotariou et al., 2010), while the global-scale GEOS-Chem model (e.g., 2° × 2.5° grid resolution) was widely used in many other studies such as those concerning the trans-Pacific export of Asian pollution (Heald et al., 2003; Walker et al., 2010). For the present study, GEOS-Chem version 9-01-01 (http://acmg.seas.harvard.edu/geos/) with the GEOS-5 assimilated meteorology was used. The original 72 vertical layers of meteorological fields were reduced to 47 layers with the lowest 4 km being resolved by 20 layers. The boundary conditions of the nested grid model domain in Asia were provided by the coarse-grid 4° × 5° global GEOS-Chem model. The model domain (10°S–50°N, 70°E–150°E) includes China, Japan, all other countries in Southeast Asia, and India. The NOₓ-Oₓ-VOCs-aerosol (full-chemistry) simulation was conducted with a spin-up run of 1-year before the actual simulation to minimize the effects of initial conditions. The GFED2 (Global Fire Emissions Database, Van der Werf et al., 2006) monthly mean bb emissions, biofuel emissions (Yevich and Logan, 2003), INTEX-B emissions [EDGAR emissions (Olivier and Berdowski, 2001) for the rest of the model domain] for combustion of fossil fuel and MEGAN model for biogenic emissions as mentioned were included in the nested model simulation.

RESULTS AND DISCUSSION

Inter-Annual Variation of CO Mixing Ratios due to Anthropogenic Emissions

To detect whether there were significant inter-annual variations of continental outflow of CO, the CO mixing ratios measured at a downwind surface-level site (i.e., Tap Mun, Hong Kong, Fig. 1) were examined. The continental outflow of anthropogenic pollution in the boundary layer peaks in March, which is the same month as when NASA’s well-known TRACE-P campaign commenced and was undertaken (Jacob et al., 2003; Liu et al., 2003). Thus the CO mixing ratios for this month in the period from 2007–2010 were selected for analysis. In order to increase the data representativeness, CO mixing ratios measured in April were also included for analysis. It is noted that April is the transition period of winter and summer, with weakened continental outflow and the effects of inflow of a cleaner marine air mass. We did not include measurements taken in February since this data is complicated by the occurrence of lower emissions during the extensive national Chinese New Year holiday period (Wai and Tanner, 2010). The measured daily average CO mixing ratios in March and
April shown in Fig. 2(a) clearly indicate that the CO mixing ratios in 2009 and 2010 are lower than those in 2007 and 2008. We also selected sampling days when there was continental outflow. These days were selected under the influence of the winter monsoon immediately after the cold front passage. We referred to the timings when Hong Kong was under the influence of the winter monsoon, as published in the weather reports by the Hong Kong Observatory. The timings were then verified by the variations of temperature and pressure, as well as by the synoptic surface pressure patterns over Hong Kong (Wai and Tanner, 2005). Each post-frontal event lasted for typically 4–5 days, which is similar to the cases reported by Wang et al. (2004). The CO mixing ratios for these sampling days feature higher mixing ratios compared with those for days without the influence of continental outflow (Fig. 3). A typical case demonstrating the CO variations before and after the continental outflow is discussed later. A comparison of CO mixing ratios for these days in 2007–08, versus 2009–10, clearly demonstrates a reduction of CO emissions at the origin of outflow (Table 1), suggesting a reduction of CO emissions (Fig. 3). The area (24.5–29.5°N, 125.5–132.0°E) is located close to one of the three highly polluted regions in China—the Yangtze River Delta (including Shanghai) (Fig. 1). The result of the MM5-CMAQ model simulation (Fig. 3(a)) demonstrates that this area is also under the influence of the continental outflow of CO. The monthly mean column CO densities in March over the area in 2007 (Fig. 2(b)) and 2008 (Fig. 2(c)) (2.38–2.80 × 10^{18} molecules/cm^2) are higher compared with those observed in 2009 (Fig. 2(d)) – 2010 (Fig. 2(e)) (2.20–2.62 × 10^{18} molecules/cm^2) with measurement uncertainty of 7–8% (Yurganov et al., 2011 and see references therein). The higher (p < 0.01) area-averaged CO densities in 2007–08 (2.50 × 10^{18} molecules/cm^2, N = 61) with respect to 2009–10 (2.32 × 10^{18} molecules/cm^2, N = 57) found herein agrees with the results from the surface-level site. Although a CO reduction was also found in the monthly results for April just as in March, they should be analyzed with caution due to the complication of pollution transport with reversed wind direction in the transition period of April. The bb plumes from Southeast Asia, which are detailed in a later Section, may have minor effects on the CO column densities due to the plume transport in the lower free troposphere. However, the inter-annual variation of the CO column densities should not be attributed to the contribution of the bb due to the inconsistency of the temporal variation.

Fig. 2. (a) Daily CO mixing ratios measured at a surface-level rural site (Tap Mun) in southern China in March and April in 2007–2010. Monthly mean CO column densities (× 10^{18} molecules/cm^2) observed by the AIRS over East China Sea and western Pacific Ocean in March in (b) 2007; (c) 2008; (d) 2009; (e) 2010.
pattern. For example, the bb-derived CO emission from Southeast Asia in 2008 is the lowest (Van der Werf et al., 2006) among the four years but the CO column densities in the year are the highest.

The global economic recession in 2008–2009 could have effects upon the reduction of ambient CO mixing ratios at the mentioned downwind locations. From MOPITT CO observations and inverse modeling, Fortems-Cheiney et al. (2011) reported that there was a sudden drop of posterior CO emissions in 2008 and a slight re-bound in 2009 in Asia. Their figures included both fossil fuel emissions in East Asia and bb emissions in Southeast Asia so that a direct application of their figures to our case is difficult. Nevertheless, these authors attributed the lower global CO emissions in 2008 and 2009 to economic recession. For other pollutants, satellite observations of aerosol optical depth and column nitrogen dioxide over northern China were found to reflect the sharp onset of the economic recession in the fall of 2008 and the rebound of the economy in the latter half of 2009 (Lin et al., 2010). Based on this fact, the reduction of observed downwind CO mixing ratios in 2009 found here could be explained, at least partially, by the reduction of CO emissions in China and the globe. However, the lower CO mixing ratios measured in 2010 are mainly due to other factor(s), of which the recent CO emissions control in China is attributed to be a major one. Further discussion on this follows. The inter-annual variation of the continental outflows is not convincing enough to explain the CO reduction. Based on the East Asia Winter Monsoon index (Jhun and Lee, 2004; Kim et al., 2013), the strength of the winter monsoon in 2008 is weaker (thus weaker outflow) than that in 2009. However, higher CO levels resulted in 2008 as discussed. The detailed discussion of the effect of inter-annual variation is out of the scope here.

To support the hypothesis that the recent change of CO emissions in China has significant effects on the downwind CO mixing ratios, the CO mixing ratios were simulated at the above-mentioned Tap Mun surface-level site by the MM5-CMAQ model. The approach is to carry out the 2007 and 2010 simulations with the meteorology of the corresponding years, but with CO emissions fixed at a base year (2006 in the present case). Then the computed CO mixing ratios are compared with the observations. When comparing the mixing ratios in 2007 and 2010, the adjustment of the CO emissions might be necessary to improve the agreement between modeling results and observations, so that one can then determine if 2007 or 2010 has higher CO emissions. This approach is basically equivalent to that adopted by Carmichael et al. (2003a) who employed a comparison between modeled values and observations in order to evaluate emissions. Wang et al. (2004) also showed that adjustment of CO emissions was required in order to improve the agreement between modeling results and observations. In the present case, since we aim to determine if 2007 or 2010 has the higher CO emissions, no re-run of the
Fig. 3. (a) Simulated surface-level CO distribution during a frontal passage over eastern Asia on 5 March 2007. The location of the cold front is arrowed; (b) Evolution of observed and modeled CO mixing ratios during the events of the continental outflow in 2007 (upper) and 2010 (lower). The timing (mm/dd/yy) of the frontal passage over Hong Kong is arrowed.

Table 1. A comparison of CO mixing ratios (ppm) measured at Tap Mun station under the influence of continental outflow in 2007–08 versus 2009–10.

<table>
<thead>
<tr>
<th>Year/Month</th>
<th>2007–08</th>
<th>2009–10</th>
<th>% reduction</th>
<th>p-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>March</td>
<td>1.05 ± 0.15 (N = 20)</td>
<td>0.44 ± 0.22 (N = 20)</td>
<td>58%</td>
<td>&lt; 0.01</td>
</tr>
<tr>
<td>April</td>
<td>1.04 ± 0.26 (N = 15)</td>
<td>0.64 ± 0.16 (N = 25)</td>
<td>38%</td>
<td>&lt; 0.01</td>
</tr>
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</table>

* Mean daily mixing ratio reported; N = number of samples.

model with adjusted CO emissions is necessary. A period of cold front passage was selected for the 2007 and 2010 simulations since the boundary-layer continental outflow features with highly-polluted continental air associated with the southeastward advection of cold fronts (Carmichael et al., 1998; Wang et al., 2004; Wai and Tanner, 2005). The scenario is visualized in Fig. 3(a).

A typical case in 2007, commencing on 4 March, was selected to demonstrate whether the MM5-CMAQ model performed satisfactory in capturing the features of the
passage of a cold front. There was a rapid drop of temperature from 21 to 10°C but an increase in surface pressure from 1010 to 1021 hPa within 2 days in Hong Kong, thereby indicated a frontal passage. The features, including the location of the front (Fig. 3(a), surface pressure map not shown), are well captured by the MM5-CMAQ model. The figure shows the computed elevated CO mixing ratio at the surface layer and the large mixing ratio gradient behind the cold front. These events are consistent with results computed by the GEOS-Chem model (Liu et al., 2003) and CFORS model (Wai and Tanner, 2005). The situation of high CO mixing ratios was maintained for ~4 days after the frontal passage, which is typical.

Fig. 3(b) shows the evolution of observed and modeled CO mixing ratios during the events of the continental outflow, featuring with a frontal passage, in 2007 and 2010. The results not only reveal the increase of CO mixing ratios after the frontal passage but also imply the change of CO emissions in China in 2007 and 2010. The CO mixing ratios for the 2007 case are substantially under-estimated compared with the 2010 case with the mean biases of -0.555 and -0.038 in 2007 and 2010, respectively. Increase of CO emissions in the model for the 2007 simulation is thus required to improve the agreement between modeling results and observations, although the performance of the meteorological model could play a minor role in the discrepancy. The results also indicate that the actual emissions which contribute to CO mixing ratios during the events of the continental outflow in China in 2007 are much higher than those in 2010 and therefore a reduction of the actual CO emissions occurred in 2010. The reduction is attributed to the CO emissions control.

Some evidence of recent reduction of CO emissions and effective control of CO in China is further discussed here. For the former, we have already demonstrated the emissions reduction by the MM5-CMAQ model simulations. Based on an inverse modeling study of CO in Eastern Asia in 2005–2010 (Yumimoto and Uno, 2012) with independent validation of emissions by satellite and surface in-situ observations, higher annual CO emissions in China were calculated in 2007 and 2008 (180.8 and 160.3 Tg/year, respectively), compared to the values in 2009 and 2010 (152.5 and 156.1 Tg/year, respectively). Through a global survey of CO observations by four satellites, a decreasing trend of CO total column was reported over north-eastern China from 2000 to 2012 (Fig. 8: Worden et al., 2013). In particular, a drop of the levels in 2009–10 compared to 2007–08 is consistent with our finding over the remote East China Sea. It was pointed out that in Beijing, there were reductions in CO emissions by 2008 (even without considering the reductions from restrictions during the Olympics) due to such measures as stricter vehicle emission standards, phasing out of residential coal stove use and changes in the industry sector. These changes in the technology mix would have a significant impact on combustion efficiency and therefore CO emissions. For the latter, the analysis of CO2 to CO ratios (Wang et al., 2010) suggested improvement in overall combustion efficiency over northern China and thus reduced CO emissions. China has implemented aggressive policies to improve energy efficiency from 2005 to 2010 (China FAQs, 2010). Other relevant studies are as follows. The CO column density data derived from AIRS revealed that there was a decreasing trend after 2007 in eastern and northern China from 2004 to 2010 (Wei et al., 2011). The decrease was due to increased combustion efficiency of coal combustion for power generation; increased usage of low-CO-emission technology (such as natural gas, hydro-power and nuclear power) for power generation; and reduced CO emissions from the steel production industry through recycling activities. In the mentioned heavily-polluted Shanghai area, the air quality in terms of PM10, SO2 and NO2 measurements in 2009 and 2010 was reported to be the best in the data set starting from 2001 (CAI-Asia, 2010; Zhen et al., 2010). The CO reduction was reported to have benefited from pollution control measures introduced for the World Expo in 2010 (http://megaevents.cleanairinitiative.org/shanghaiexpo2010/air%20Quality%20measures%20in%20Shanghai).

### Contribution of Biomass Burning to CO Mixing Ratios in the Free Troposphere

To better understand the influence of bb from Southeast Asia upon the downwind area, satellite retrievals were used to examine the bb sources and to detect CO enhancements in downwind areas, with further insights given by the chemical transport modeling results. Based on the MODIS 10-day fire maps, active bbs were found at the western coast (near Bay of Bengal) and eastern inland (near the boundaries with China and Thailand) areas of Myanmar, northern Thailand and part of southern Vietnam in March 2008. There was a northward shift of the bbs in April 2008. The locations and emissions are generally consistent with the monthly GFED2 data.

The high-resolution model was employed for scenarios when there were high bb impacts on downwind southern China and the Western Pacific Ocean. Both TES and MOPITT retrievals were used for different purposes. The CO episodes were firstly selected using the TES level-3 daily global CO mixing ratios at 681 hPa (horizontal resolution of 2° latitude by 4° longitude), with mean CO mixing ratios higher than 170 ppb over a downwind area. The area (115°E–124°E, 20°N–26°N) covers the Western Pacific Ocean, Taiwan, and part of southern China. It comprised eight episodes in March and April 2008 which were selected for analysis. Free-tropospheric CO mixing ratios were selected since the bb-derived CO, which is discussed in more detail later, contributes significantly to the total mixing ratio. With higher resolution as mentioned, the MOPITT retrievals at 700 hPa for the episodes were then used for comparison with the modeling results at the same level (Fig. 4(a)).

Due to the limited area coverage per overpass and missing data of the daily MOPITT CO retrievals, the detailed comparison between observed and modeled CO distributions for the whole study area is not possible. Only the CO satellite retrieval with an area at least $2^\circ \times 2^\circ$ was sampled and compared with the modeling results. The sampled CO retrievals were also selected at different locations and mixing ratios to reduce bias of the comparison. Fig. 4(a)
Fig. 4. (a) Scatter plot of simulated CO mixing ratios by GEOS-Chem model versus observed CO mixing ratios by MOPITT ($N = 24$). Case study on 22 March 2008; (b) MODIS satellite image showing intensive burning events over southeast Asia (in black square), with dense cloud clusters to the northeast; (c) altitude-longitude plot of simulated CO mixing ratios at latitude 21°N; and (d) simulated burning-derived CO distribution over southeast Asia, South China Sea and the western Pacific Ocean.
shows a scatter plot of modeled CO versus observed CO mixing ratios with slope of the best fit line of 0.93 ($r^2 = 0.72$), with performance statistics: normalized mean bias (NMB) = −7%, normalized mean error (NME) = 13% and root mean square error (RMSE) = 34.3 ppb, suggesting relatively good agreement of the modeling results with observations when there were high bb impacts on the downwind areas.

Within the episodes, the modeled downwind CO mixing ratios at 700 hPa over the western Pacific Ocean (25°N–30°N, 130°E) reached up to 200 ppb. This value is comparable to those observed and simulated by a coarser-grid model during the TRACE-P campaign (Liu et al., 2003). Separate model runs with and without the bb emissions were made to calculate the contribution of bb-derived CO in the lower free troposphere. The difference between the two runs gives the contribution of the bb. Within the episodes, the maximum contribution by the bb to the total CO mixing ratio at 700 hPa ranges from 33 to 86% over the downwind South China Sea and western Pacific Ocean, pointing out the importance of the bb contribution to modify the tropospheric CO in the lower free troposphere. The bb-derived CO plumes for all episodes contribute up to 100 ppb at 700 hPa over the downwind area, comparable to the results reported elsewhere (Carmichael et al., 2003b; Liu et al., 2003). Additionally, our high-resolution model can better resolve the plume structure and its field of mixing ratios.

While convective export of the bb plume and its outflow have not been studied in detail by a combination of satellite image, high-resolution chemical transport model computation and surface pressure map, herein a case study was undertaken by using these tools. The MODIS satellite image showed intensive bb events over Myanmar on 22 March 2008 (Fig. 4(b)). While smoke was clearly seen over the large-scale bb area, dense cloud clusters were also located to the northeast, denoting that active convective activities prevailed in that month. The convection was also supported by low pressure systems (1008 hPa) shown in the surface pressure map within the area (figure not shown). An altitude-longitude plot at 21°N (Fig. 4(c)) shows relatively uniform high CO mixing ratios (~ 180 ppb) from the ground to 3 km a.s.l., indicating rapid vertical transport of the bb plume from the boundary layer to lower free troposphere. The tendency of eastward plume transport is evident from the altitude-longitude plot and computed bb-derived CO distribution at 700 hPa (Figs. 4(c) and 4(d)), since the westerly dominated in the free troposphere. It is noted that zonal transport of the bb plumes with elevated CO mixing ratios to the Western Pacific Ocean in March and April in the lower free troposphere are common phenomena, based on the modeling results. For the case on 22 March, the bb-derived CO plume was over the Western Pacific Ocean (longitude ~127°E) of 70 ppb (or 35% of the total) at 700 hPa (Fig. 4(d)).

CONCLUSIONS

In this study, the springtime inter-annual variations of CO over the rural location Tap-Mun, Hong Kong and the continental outflow to the western Pacific based on CO column density measurements by satellite for the period 2007–10 have been discussed. A decrease in surface CO at the measurement location and outflow region during 2009–10 was found, which is attributed to the pollution control measures taken and global economic recession. The emission reduction was supported by numerical model simulation with CO emissions fixed to 2006 levels. Since the free troposphere over southern China is also influenced by bb in Southeast Asia during springtime, the second aspect discussed in the paper is the contribution of bb derived CO to the free troposphere by selecting some episodes of high CO levels. The major findings are summarized below:

• The CO mixing ratios measured at a surface-level rural site under the influence of continental outflow in March and April showed substantial reductions (at 58% and 38%, based on the 2007–08 level, respectively) in 2009–10;
• The monthly mean column CO densities in March derived from the AIRS over the East China Sea adjacent to the Western Pacific Ocean showed the same trend as the results measured at the surface-level site, suggesting that CO reduction was a regional phenomenon;
• The reduction of observed downwind CO mixing ratios and column densities in 2009 and 2010 is attributed to the reduction of CO emissions in China and the global economic recession. Evidence of recent improvement of combustion efficiency and effectiveness of CO emissions control in China have been discussed;
• The surface-level CO mixing ratios at the rural site simulated by the MM5-CMAQ model not only demonstrated the model capability in capturing the evolution of CO mixing ratios during the frontal passage but also supported the change of CO emissions in continental China in 2007 and 2010;
• With the GFED2 bb emissions inventory, the high-resolution nested-grid GEOS-Chem model computed CO mixing ratios at the lower free troposphere which were in agreement with those observed by the satellite, during the selected episodes;
• Within the episodes, the maximum contribution by bb to total CO mixing ratio at 700 hPa ranged from 33 to 86% over the downwind South China Sea and western Pacific Ocean, pointing out the importance of the bb contribution to modify the tropospheric CO level in the lower free troposphere;
• The bb event on 22 March 2008 has been studied and interpreted in detail by a combination of tools including MODIS image, chemical transport models and surface pressure map.

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