Simulating the transport and chemical evolution of biomass burning pollutants originating from Southeast Asia during 7-SEAS/2010 Dongsha experiment

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**HIGHLIGHTS**
- Reanalysis of transport of biomass burning plume originating from SEA.
- Chemical evolution of biomass burning pollutants during long-range transport.
- Combine WRF/HYSPLIT/CMAQ to analyze the compositions of biomass burning plume.

**ABSTRACT**
This study aimed to simulate the transport of biomass burning (BB) aerosol originating from Southeast Asia (SEA) during the Dongsha Experiment conducted from March 2010 to April 2010. Transport pathways were reanalyzed and steering flow in the mid-latitude areas and anticyclones in low-latitude areas were found to control the transport of BB plume after it was injected to a high atmosphere. For the 12 simulated and observed events at Mt. Lulin (2862 m MSL; 23°28′07″ N, 120°52′25″ E), the 72 h backward trajectories were all tracked back to southern China and northern Indochina, which were the locations of the largest BB fire activities in SEA. Chemical evolutions of BB pollutants along the moving trajectories showed that organic matter was always the dominant component in PM2.5, consistent with the observations at both near-source regions and Mt. Lulin. For nitrogen species, nearly all NOx molecules oxidized into HNO3, NO2, PAN, and PANX in fires or near fires. The synchronic consumption of NOx, SO2, and NH3 explained the production of the major components of inorganic salts. In the moving BB plume, sulfate concentration increased with decreased nitrate concentration. Ratios of ammonium to PM2.5 and elemental carbon to PM2.5 remained nearly constant because additional sources were lacking.

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1. Introduction

Biomass burning (BB) is generally recognized as an important factor that can influence global or regional carbon cycle, meteorology, hydrological cycle, radiative budget, and even climate change. For example, massive amounts of black carbon (BC) in BB aerosols absorb solar radiation and warm the atmosphere of the
winds that westerly winds mainly concentrated in SEA and southern China. In April 2006, the
ence on the effect of BB on the Yangtze River Delta region between
range transport on downwind regions, including the Pearl River
model to assess the effect of BB aerosols from SEA on East Asia (EA).
explained the same event further and suggested that BB in SEA
plume from SEA to Taiwan using two examples. This study
provides information about the chemical characteristics of BB
plumes, which could further explain optical characteristics and
cloud physics for future studies.

2. Methods

2.1. Model description and configuration

The principal modeling tools and resources of the present study are based on those that Fu et al. (2012) and Huang et al. (2013) used. CMAQ model (Byun and Schere, 2006) was used to simulate the chemical field during the 2010 Dongsha experiment from March 2010 to April 2010. The inputs for the CMAQ model included meteorological data from the simulation results of the WRF model (Wang et al., 2012) and various emissions. Anthropogenic emission was based on the 2006 INTEX-B (Intercontinental Chemical Transport Experiment-Phase B) emission inventory of NASA (Zhang et al., 2009). This basis may underestimate the simulated results because the growth of anthropogenic activities from 2006 to 2010 was ignored. Biogenic emissions were generated using the MEGAN (Model of Emission of Gases and Aerosol from Nature) model (Guenther et al., 2012). This model focused on BB emissions from the FLAMBE project (the joint Navy, NASA, NOAA, and Universities Fire Locating and Modeling of Burning Emissions, Reid et al., 2009). Fu et al. (2012) provided further details of model configurations. However, the present study made several changes. For example, the simulation range was changed to three nested domains with res-
olutions for domains 1, 2, and 3 at 45, 15, and 5 km, respectively (Fig. 1). The simulation results of the second domain were analyzed for chemical evolution along the long-range transport from SEA to
Taiwan; the third domain was used for the comparison with the observations at Mt. Lulin in Taiwan. The outputs of the WRF simulation were used for the HYSPLIT model simulation to obtain the historical trajectories of the BB plumes that arrived at Mt. Lulin. From the simulation results, we obtain the BB plumes and chemical compositions at the locations for each hour.

2.2. Uncertainty of BB emissions

Several studies have discussed the uncertainty of BB emissions and indicated the difficult-to-avoid inaccuracies of data, such as burned area, burned time, fuel loading, burning efficiency, and

Fig. 1. Three nested domains for current simulation.
burning state, which may lead to uncertainty during emission estimation (Campbell et al., 2007; van der Werf et al., 2006, 2010; French et al., 2011; Zhang et al., 2012). Although several well-known multi-compositions of BB emission inventories exist, which are widely used and readily downloadable (for example, FLAMBE: Reid et al., 2009; GFEDv4: Giglio et al., 2013; IS4FIRES: Sofllev et al., 2009. FINN: Wiedinmyer et al., 2011; RCP: van Vuuren et al., 2011), these projects cannot determine the most feasible inventory in all regions around the world or in all periods at all times. The present study followed the previous studies (Fu et al., 2012; Huang et al., 2013) and applied the FLAMBE BB inventory, which also met difficulties. Fu et al. (2012) suggested that CO emission in FLAMBE is probably overestimated in Myanmar and major areas of Thailand, thereby causing an overestimation of CO concentration during peak episodes. They also compared the BB emission inventories for both FLAMBE and GFEDv2 (van der Werf et al., 2006) and found that the former is 7.89 and 11.63 times higher than the latter in March and April 2006 in SEA, respectively. Fu et al. (2012) selected the FLAMBE emission inventory in their study for three reasons. First, the FLAMBE performs better than GFED in terms of simulation with observation at the NASA-owned temporary site in South Thailand. Second, the FLAMBE has an hourly temporal profile. Third, Fu et al. (2012) adopted the suggestion of Nam et al. (2010), in which the GFED emission inventory may underestimate CO at relatively low subtropical latitudes over Asia. Huang et al. (2013) also found the BB emission in the 2006 FLAMBE is overestimated over Myanmar and major areas of Thailand. The criticism about the overestimation of FLAMBE was also suggested in Reid et al. (2009). Therefore, the FLAMBE emission inventory could likely overestimate northern Indochina, which is the main BB source region for downwind areas, including Taiwan.

To increase the quality of quantification of the simulation results, a sensitivity test for the FLAMBE emissions was performed. The BB emission data for all BB emission pollutants were multiplied with 1.0, 0.5, 0.25, 0.125, and 0.0, which were then used as inputs for the simulations. Compared with the continuously monitored CO (Ou-Yang et al., 2014), O3 (Ou-Yang et al., 2012), and PM10 data at Mt. Lulin (Fig. 2) that Taiwan EPA managed, the original BB emission intensity that was multiplied with 0.25 was the most feasible in terms of various statistics, such as RMSE (Root Mean Square Error), MNB (Mean Normalizes Bias), MNE (Mean Normalized Error), MFB (Mean Fractional Bias), MFE (Mean Fractional Error), IOA (Index Of Agreement), F2 (Factor 2 analysis), and R (Correlation Coefficient) (Table 1). Therefore, all BB emissions were reduced by 75% for further analysis. The IOAs of CO, O3, and PM10 are all above 0.6 (Table 1), which indicates a relatively good performance. The purpose of reducing the intensity of BB emissions in the FLAMBE emission inventory was to simulate the scenes and not to tune satisfactory results. However, reducing emission may also compensate for other systematic biases in the modeling system. Compared with the observed concentrations at Mt. Lulin during 7-SEAS/Dongsha Experiment (Lin et al., 2013), the simulated and observed means of PM2.5, SO2, NH3, NO3, OC, and EC at Mt. Lulin were 13.7 μg m−3 and 14.2 μg m−3, 3.2 μg m−3 and 2.3 μg m−3, 1.7 μg m−3 and 0.9 μg m−3, 2.4 μg m−3 and 0.6 μg m−3, 4.8 μg m−3, and 4.6 μg m−3, 0.5 μg m−3 and 0.8 μg m−3, respectively. These simulation results agreed well with the observations except for NO3, which were underestimated. As explained later in section 3.2, the condensation of HNO3 that arose from the drop of temperature when the BB plume approached Mt. Lulin caused the overestimation.

2.3. BB events detected at Mt. Lulin

Given that PM10 > 35 μg m−3 and CO > 0.3 ppm was observed at Mt. Lulin as the standard of events, 12 events were observed and simulated from March 2010 to April 2010 (Fig. 2). These 12 events peaked at Mt. Lulin on March 12, 03:00 h (LST); March 12, 16:00 h; March 16, 18:00 h; March 18, 01:00 h; March 21, 08:00 h; March 25, 17:00 h; March 26, 05:00 h; March 28, 06:00 h; April 5, 19:00 h; April 6, 19:00 h; April 11, 03:00 h; and April 11, 16:00 h. The 72 h backward trajectories starting from Mt. Lulin at the peak hour for

Fig. 2. The comparison of observed and simulated CO, O3, and PM10 at Mt. Lulin site for the BB emission data multiplied with 1.0, 0.25, and 0.0, respectively. Two lines with BB emission data multiplied with 0.5 and 0.125 were skipped but can be imaged to exist between 1.0 and 0.25 and between 0.25 and 0.0.
the 12 events are illustrated in Fig. 3. As expected, all of the 12 trajectories traced back through southern China/northern South China Sea and to northern/central Indochina because the BB plume that originated from Indochina affected the Mt. Lulin site (Lee et al., 2011; Chuang et al., 2014). Ensemble backward trajectories have also been conducted (not shown), and the 12 trajectories (Fig. 3) have been proven representative. The trajectories between the source region and Taiwan appeared as straight lines, which indicate the shortest path between these two locations (Fig. 3). After the BB plumes were injected into a high altitude, these plumes arrived at Taiwan in approximately 2–2.5 days. This finding implies that the BB plumes arrived in 2–2.5 days and maintained concentrations as high as possible from SEA to Mt. Lin. Although the 12 events were both simulated and observed during the 2010 Dongsha campaign, only two representative events were discussed in the present study. One event occurred in March and the other in April 2010. The rest of the events have similar transport pathways, and the chemical evolution of the BB pollutants to these two events was not repeated.

3. Results and discussion

3.1. Transport pathways of BB plumes

In low-latitude equatorial areas, thermal buoyancy and deep thermal convection, such as the inter-tropical convergence zone (ITCZ) could explain the lifting of BB pollutants (Folkins et al., 1997). However, the thermal convection over Indochina is not as strong as that around the equator and cannot explain the uplifting of BB pollutants. Therefore, other factors help uplift BB pollutants from near the surface to high altitudes, as suggested in previous publications.

Several studies have indicated that BB plumes emerging around northern Indochina would ascend to a high atmosphere and then move eastward using the westerly winds (Lin et al., 2009, 2013; Yen et al., 2013). The movement of BB plumes originating from SEA suggests that at least two factors can determine if BB plumes would pass Mt. Lulin. The first factor implies whether forces can help the BB plume rise up above the PBL around the northern/central Indochina. This mechanism includes orographic lifting, the upward motion in low-atmospheric low or a strong updraft in the lee vortex. The second factor implies whether the air stream in the westerly winds can transport the BB plume to Mt. Lulin. Actually, the transport pathways of BB plumes depend on the steering flow in middle-latitude areas and the anticyclone in low-latitude areas over the source and the downwind regions (Fig. 4). Given that the 925 hPa to 700 hPa trough and the BB source region usually overlap over Indochina, the wind fields at the post-trough downwind areas are usually westerly/southwesterly winds. If the BB plumes transport along the southwesterly winds, then the BB plumes may transport northeastward to middle/high-latitude regions. If the wind direction is from the west, then the BB plumes may transport eastward and sometimes transport into the anticyclone in low-latitude areas. For example, on March 15, 2010, most parts of the BB plume followed the steering flow and transported northeastward and only a small part at the south edge was pulled into the anticyclone at low-latitude areas (around two places at 128°E, 20°N and 110°E, 17°N) (Figs. 4a and 5a). Given that the main stream of BB plumes moved northeastward, Taiwan was located at the south edge of the BB plume. When the range of influence from the anticyclone became large, more BB pollutants were transported into the anticyclone (Figs. 4b and 5b). On March 17, 2010, Taiwan was located in the middle of the eastward-moving BB plume. If the range of influence from the anticyclone over the Pacific shrinks (Fig. 4c) and the flow field at the east flank of the trough becomes parallel with line of 30°N, then the BB plume also becomes parallel with 30°N (Fig. 5c). This weather pattern mostly prevents the BB plume from moving to high-latitude areas and only allows movement in low-latitude areas. Therefore, the BB plume may move eastward and pass through south of Taiwan to the Pacific.

In section 2.2, this study has used PM_{2.5} to validate the model performance. In the following, this study used PM_{2.5} to illustrate the chemical evolution of aerosols in the BB plume. Although PM_{2.5} at Mt. Lulin has no continuous measurement, PM_{10} can be used to replace PM_{2.5} for modeling validation because the PM_{2.5}/PM_{10} ratios were nearly united during BB season (Lin et al., 2013). Furthermore, the simulated mean of PM_{10} from March 1 to April 30 2010 was 13.7 μg m\(^{-3}\), which was slightly lower than the measured
mean of PM$_{2.5}$, which was 17.5 $\mu$g m$^{-3}$ for the BB events from 2003 to 2009 (Lee et al., 2011). The simulation results should be reasonable because the simulation period also include some non-BB events.

### 3.2. Chemical evolution of BB pollutants in BB plume

HYSPLIT model was applied to determine the backward trajectory of BB parcel starting from the hour when a peak concentration occurred at Mt. Lulin to the preceding 72 h. Thus, we can understand the chemical evolution of BB pollutants in the BB parcel in the last three days before the BB parcel arrived at Mt. Lulin. However, a peak concentration at Mt. Lulin does not guarantee that the peak concentrations of BB pollutants would always remain at a high level along the backward trajectory because the air near the trajectory can influence the air along the trajectory (i.e., mixing). The air near the trajectory could come from a clean background atmosphere, a new BB plume, an aged BB plume, and a polluted atmosphere from Asia or local pollution. Moreover, the 12 trajectories (Fig. 3) were...
3.2.1. BB event on March 12, 2010, 16:00 h

Given that the westerly winds transported the BB plumes horizontally after lifting above the boundary layer, we selected layer 10 among the vertical layers, which has nearest the height of Mt. Lulin (2862 m), to explain the horizontal transport of BB plumes. The PM$_{2.5}$ concentration was approximately 60 μg m$^{-3}$ to 70 μg m$^{-3}$ in the BB parcel (19° N, 120° E) at approximately 3 km aloft and around southern Taiwan on March 10, 2010 (Fig. 6a). At this moment, Taiwan was located north of the BB plume, and the prevailing winds were northwesterly winds. Both the observed PM$_{10}$ (Fig. 2) and simulated PM$_{2.5}$ concentrations at Mt. Lulin were approximately 15 μg m$^{-3}$ to 20 μg m$^{-3}$ because the BB plume did not affect Mt. Lulin. Once the trough became less obvious and the low-latitude anticyclone (14° N to 20° N, 118° E to 135° E) became evident, the northwesterly became westerly winds over the South China Sea (SCS), the local high concentration of BB plume over southern China (dist N, 106° E) moved to SCS, and the foregoing BB plume gradually influenced Mt. Lulin. The observed PM$_{10}$ (Fig. 2) and simulated PM$_{2.5}$ concentrations on March 11, 2010 were approximately 30 μg m$^{-3}$ at Mt. Lulin (Fig. 6b). On March 12, 2010, the westerly winds became southwesterly winds aloft western Taiwan (23° N, 115° E), and part of the concentrated BB plume moved northeastward and passed Mt. Lulin, which observed PM$_{10}$ (Fig. 2) and simulated PM$_{2.5}$ rose to higher than 40 μg m$^{-3}$ (Fig. 6c).

The vertical section A–A (Fig. 6a–c) is indicated in Fig. 7a–c daily at 08:00 h from March 10, 2010 to March 12, 2010. Two intensive fires were located at the mountains of Myanmar (93° E to 95° E) and the border of Myanmar and north of Thailand (97° E to 99° E) below 5 km on March 10, 2010 (Fig. 7a). These two dense plumes were not obvious in layer 10 (Fig. 6a), which was approximately 3 km in height because the PM$_{2.5}$ concentrations reduced rapidly above 3 km. However, an apparent BB plume concentrated at a height of 2.5 km–4 km was located aloft the border of China and Vietnam (104° E to 112° E) (Figs. 6a and 7a). On March 11 at 16:00 h, the BB plume popped out at the border of Myanmar and northern Thailand merged with the downwind plume aloft the border of China and Vietnam, and then moved eastward (Figs. 6b and 7b). On March 12 at 16:00 h, the merged plume ranged from 1.5 km to nearly 5 km vertically and continued to move eastward, which gradually influenced Mt. Lulin (Figs. 6c and 7c). The BB plume continued to move eastward, and the simulated PM$_{2.5}$ concentration at Mt. Lulin increased to above 50 μg m$^{-3}$. Basically, the simulation result is satisfactorily accepted. The simulated PM$_{2.5}$ at Mt. Lulin was above 30 μg m$^{-3}$ on March 11 at 21:00 h, peaked to 53 μg m$^{-3}$ at on March 12 at 3:00 h, and then decreased below 40 μg m$^{-3}$ until March 12 at 17:00 h. The PM$_{10}$ concentration was above 30 μg m$^{-3}$ since 03:00 h on March 11, peaked to 64 μg m$^{-3}$ at on March 12 at 1:00 h, and then decreased below 40 μg m$^{-3}$ until March 12 at 12:00 h (Fig. 2).

Exploring the chemical evolution of BB pollutants during long-range transport is interesting (16:00 h on March 10, 2010 to 16:00 h on March 12, 2010). The air parcel that originated from Myanmar passed the border of China and Liao and the border of China and Vietnam before arriving at Mt. Lulin. Compared with the distribution of fire figures and emission inventory (not shown), the air parcel can be considered to be part of the BB plume. The BB-related chemical species and their variation in the BB parcel were evaluated for every hour during the aforementioned three-day trajectory. The air parcel that was initialized on March 10 at
The simulated PM$_{2.5}$ contour of the vertical section A–A in Fig. 6(a) at daily 16:00 h on (a) March 10 2010, (b) March 11 2010, and (c) March 12 2010, respectively.

From the position at which the center of the BB parcel was at 02:00 h on March 11 to Mt. Lulin, the NO$_x$, NH$_4$NO$_3$, NO$_3^-$, NH$_4$$_2$SO$_4$, and many chemical species slightly decreased with the distance related to dilution (Fig. 8b). Primary gases, including SO$_2$, NO$_x$, and NH$_3$ decreased to nearly zero because they were converted into the aerosol phase (Fig. 8c and d). The Sulfur Oxidation Ratio (SOR, ranging from 0.0 to 1.0, the minimum and maximum indicates that sulfur exists in the primary state [SO$_2$] and secondary state [SO$_4^{2-}$], respectively; Fan et al., 2015) in the BB parcel remained from 0.5 to 0.6 over the source regions and increased to be above 0.6 until the BB parcel moved away from source regions (Fig. 9a). Relatively, the proportion of NO$_3^-$ and NH$_4$ in PM$_{2.5}$ remained nearly constant without affecting dilution with distance (Fig. 8f). On March 12 at 1:00 h, the proportion of NO$_3^-$ in PM$_{2.5}$ decreased but increased again when approaching Mt. Lulin because of condensation. However, the condensation of HNO$_3$ and formation of NO$_3^-$ could have been overestimated because NO$_3^-$ was overestimated when compared with observations (section 2.2). From Fig. 8a, c, and f, the drop of temperature in the BB plume when approaching Mt. Lulin caused the condensation of HNO$_3$. By contrast, SO$_4^{2-}$ in PM$_{2.5}$ increased (Fig. 8f). Meanwhile, the SOR increased be nearly 1.0 (Fig. 9a). The air parcel also moved over SCS and descended, and then ambient temperature increased. Part of NH$_4$NO$_3$ evaporated, and the evaporated NH$_3$ reacted with H$_2$SO$_4$, which was produced from the conversion of available SO$_2$ (SO$_2$ reduced to nearly zero after 01:00 h on March 12 in Fig. 8d). The trend of organic matter (OM) is similar to PM$_{10}$ or PM$_{2.5}$ (Fig. 8e and b) because the proportion of OM in PM$_{2.5}$ was the highest among all aerosol compositions (Fig. 8f). This phenomenon is consistent with observations at both near-sources regions (Chuang et al., 2013) and Mt. Lulin (Chuang et al., 2014). The trend of OM and EC showed that most of the OM was primary OM. In this simulation study, which involves different primary and secondary OM sources, the primary OM from the anthropogenic (AORGPAT variable in model outputs) sources were dominant, which accounts for more than 90%. Although primary organic carbon (POC) is more than secondary OC for the SEA BB aerosols sampled at Mt. Lulin (Chuang et al., 2014), the proportion of primary OM from anthropogenic sources could be overestimated in present simulations. This result can be attributed to the possible overestimation of the emission factor of POC in the study by Andreae and Merlet (2001), which is also used in the present study.

Robinson et al. (2007) conducted a chamber experiment that revealed substantial evaporation of primary organic aerosol (POA) when very high-temperature emissions are diluted into the
atmosphere. Most of the gaseous volatile organic compounds would condense back to particles after the environment is cooled (Jimenez et al., 2009). In the experiment of Andreae and Merlet (2001), the condensed OM was attributed to the POA. However, the semi-volatile and intermediate-volatility organic compounds that evaporated from POA may form SOA through oxidation from OH attack before condensation. Therefore, the emission factor of POC could be overestimated. By contrast, SOA could be underestimated. For aging aerosols, water-soluble OC (WSOC)/secondary organic aerosol (SOA) is usually produced during long-range transport (Aggarwal and Kawamura, 2009). However, the production of SOA was not obvious in the present simulation. This result

Fig. 8. The chemical evolution of meteorological variables and chemical species in the BB parcel which arrived at Mt. Lulin at 16:00 h on March 12, 2010 (Temp: temperature; QV: water vapor mixing ratio; PAN: peroxo acetyl nitrate; PANX: peroxy propionyl nitrate (PPN) + peroxymethacryloyl nitrate (MPAN); AORGAT: anthropogenic primary organic aerosol; AORGAT: anthropogenic secondary organic aerosol; AORGAT: biogenic secondary organic aerosol; AORGCT: cloud secondary organic aerosol).

Fig. 9. The variation of sulfur oxidation ratio (SOR) and nitrogen oxidation ratio (NOR) in the BB parcel which arrived at Mt. Lulin (a) at 16:00 h on March 12, 2010 and (b) at 19:00 h on April 6, 2010.
implies that the organic chemistry in CMAQ needs improvement, particularly in the forming SOA. For example, OC may evaporate and oxidize to WSOC and condense on aerosol during long-range transport (Aggarwal and Kawamura, 2009).

### 3.2.2. BB event on April 6, 2010, 19:00 h

Three days before the occurrence of peak concentration, on April 6, 2010 at 19:00 h, Taiwan was already in the distribution range of the BB plume (Fig. 10a). On April 4, 2010, the simulated PM$_{2.5}$ was approximately between 10 μg m$^{-3}$ and 35 μg m$^{-3}$ at Mt. Lulin (Fig. 10a), which was satisfactory because the observed PM$_{10}$ was between 18 μg m$^{-3}$ and 37 μg m$^{-3}$ (Fig. 2). Although the two high-concentration BB puffs centered over north of Indochina and Hainan, both were still far from Mt. Lulin, and the foregoing BB concentration BB puffs centered north of Indochina and 120°E, 2 km further and covered the whole of Taiwan (northern Vietnam, and the BB sources, the air parcel began to descend, and the ambient anticyclone. The overestimated simulated PM$_{2.5}$ moved northeastward, and another part continued to move into the Bashi Channel. Part of the BB pollutants passed Taiwan and the SCS gradually merged into one elongated BB plume and passed the SCS and the Bashi Channel toward the Pacific. This result is ascribed to the moving direction of the BB plume that was closely connected to the traction of the anticyclone over the SCS, Philippine Sea, and the Pacific Ocean. On April 5, the two large above mentioned BB puffs moved slowly, but only part of the BB puffed over Hainan had rapidly moved to the Bashi Channel (Fig. 10b). This movement caused the overestimation of PM$_{2.5}$ (30 μg m$^{-3}$ to 60 μg m$^{-3}$) at Mt. Lulin and the observed PM$_{10}$ was only 20 μg m$^{-3}$ to 45 μg m$^{-3}$. Part of the BB plume transported to low latitudes because the traction of the anticyclone was obvious on April 6 (Fig. 9c). Similar to the first case discussed in the previous section, the anticyclone in the low latitude served an important function in inhibiting the BB pollutants that spread to high-latitude areas. On April 6, the two BB puffs gradually merged into one elongated BB plume and passed the SCS and the Bashi Channel. Part of the BB pollutants passed Taiwan and moved northeastward, and another part continued to flow into the anticyclone. The overestimated simulated PM$_{2.5}$ and observed PM$_{10}$ were 30 μg m$^{-3}$ to 70 μg m$^{-3}$ (Fig. 10c) and 20 μg m$^{-3}$ to 40 μg m$^{-3}$, respectively. Although the PM$_{2.5}$ concentration at Mt. Lulin was overestimated, the trends of the simulated PM$_{2.5}$ and observed PM$_{10}$ were similar (Fig. 2).

![Image](Fig. 10. The simulated PM$_{2.5}$ contour and wind field at layer 10 at daily 19:00 h on (a) April 4, 2010, (b) April 5, 2010, and (c) April 6, 2010, respectively.)

The vertical sections A–A (Fig. 10a–c) are indicated in Fig. 11a–c at 19:00 h daily from April 4, 2010 to April 6, 2010. On April 4, the BB plume originated from the mountains in northern Vietnam (Fig. 11a). However, the intensity of the BB plume is not as strong as those originating from the mountains in Myanmar and in the border of Myanmar and north of Thailand (Fig. 11c, Fig. 7a–c). Therefore, the BB plume did not eject vertically but rose and moved eastward simultaneously (Fig. 11b, BB puff centered at 112°E, 2 km). On April 5, the foregoing BB plume continued to influence Mt. Lulin and decelerated because the anticyclone caused the BB plume to change direction (Fig. 11b). On April 6, the leading edge of the above mentioned BB puff centered at 112°E, 2 km further and covered the whole of Taiwan (Fig. 11c, BB puff centered at 120°E, 2.5 km).

On April 3, 2010, the BB parcel was aloft northern Laos and northern Vietnam, and the PM$_{2.5}$ concentration in the parcel could reach 200 μg m$^{-3}$ (Fig. 12b). After the BB parcel moved away from the BB sources, the air parcel began to descend, and the ambient atmospheric pressure and ambient temperature gradually increased (Fig. 12a). All of the concentrations of major BB pollutants decreased because of the dilution with the transport distance (Fig. 12b). After the BB parcel moved away from the fire sources, NO$_x$ in the air parcel was consumed, and PAN, PANX, and HNO$_3$ were produced instead (Fig. 12c). As illustrated in Fig. 9b, the NOR increased to be over 0.5 at the source regions. The NOR at the source regions is lower than that (above 0.9) for the March 12 event because the NO$_x$ concentration was high for this event. However, the NOR still increased to be above 0.9 soon after the BB parcel was away from the source regions (Fig. 9b). The SO$_2$ concentration in
this event was also higher than that in the March 12 event (Fig. 12d). The SOR was relatively lower than the source regions, which is approximately 0.3 (Fig. 9b). On April 4 at 23:00 h, the BB parcel that moved eastward over Hainan suddenly fell approximately 500 m, and the ambient temperature increased to approximately 5 °C. This movement decreased the amounts of NO$_3^-$ and NH$_4^+$ to 8.8 μg m$^{-3}$ and 3.8 μg m$^{-3}$, respectively. HNO$_3$ and NH$_3$ also increased to approximately 2.2 ppb and 0.9 ppb, respectively. On April 5, the trends of NH$_3$ and HNO$_3$ in PM$_{2.5}$ exhibited an opposite trend (Fig. 12f). The PAN continuously decreased during the transport because of the dilution and conversion to the aerosol phase. Meanwhile, SO$_2$ concentrations continuously decreased because of oxidation and diffusion (Fig. 12d). Relatively, the percentage of SO$_4^{2-}$ in PM$_{2.5}$ gradually increased (Fig. 12f). Hereafter, the BB parcel moved northeastward and continued to mix with clean air from the SCS. The concentrations of major BB pollutants, such as PM$_{10}$/PM$_{2.5}$, OM, EC, and CO, gradually decreased until approximately 12 h before reaching Mt. Lulin (Fig. 12b and e). Most of the BB pollutants were enhanced when the BB plume approached Mt. Lulin. The moving air that was blocked and squeezed in the windward side of the Central Mountain Range did not influence the enhancement. Instead, pollutants from Asia influenced the enhancement. This result implied that SO$_2$ was also enhanced but was negligible in fresh BB plume. Once the SO$_2$ in the fresh BB plume was consumed, its concentration should remain at approximately zero. During the long-range transport, the variation of SULF is in wave form because SULF was mainly produced through gas-phase reaction of SO$_2$ and OH, which usually dominates at noon (Stockwell and Calvert, 1983). In this case, the aqueous-phase reaction for the production of SULF was insignificant because the peak of H$_2$O$_2$ was behind the peak of SULF (Fig. 12a), and the water mixing ratio remained constantly low (Fig. 12a) (Kleinman, 1991). Since the second day, the trends of SO$_2$ and H$_2$O$_2$ were nearly the same. This finding indicates that the BB parcel was not found in the environment-like clouds or fogs (Pandis and Seinfeld, 1989). Similar to the March 12 event, the proportion of OM in PM$_{2.5}$ was the highest among all aerosol compositions (Fig. 12f). Meanwhile, the trend of EC, the anthropogenic POA, and OM were similar (Fig. 12e). This result implies that the primary OM could be overestimated and relatively, the SOA is underestimated. Therefore, the evaporation of VOCs from POA and the oxidation and condensation of oxidized VOCs to form SOA were eager to be discovered and incorporated into air-quality models.

If the bottom of the plume is below the heights of the mountains in Taiwan, then the air quality on the approaching BB plumes could influence the air quality on the ground. Yen et al. (2013) explained that the downward airstream along with the passage of the front could bring high-elevation BB plume to the ground. An explanation to this situation is worth studying. In addition, the chemical reactions of BB pollutants and the locally emitted pollutants should produce few secondary products because the precursor gas concentrations in the BB plumes of the above mentioned events were low when approaching Taiwan. This hypothesis must be verified and the detailed discussion of the BB plume approaching Taiwan will be discussed in the near future.

4. Conclusions

This study is part of the 2010 Dongsha Experiment and a pre-study of the 7-SEAS project. The WRF and CMAQ modeling system was used to simulate the transport and chemical evolution of BB plume from source regions to the Mt. Lulin site in Taiwan.

Our simulation results reanalyzed the transport of BB plumes and found that after the BB pollutants were injected to high...
altitudes, the steering flow in middle latitude areas and anticyclone in low-latitude areas controlled the transport of BB plumes. If the BB plumes were transported along the southwesterly winds, then the BB plumes may transport northeastward to the middle/high-latitude regions. If the BB plume transported along the westerly winds, then the BB plumes may transport eastward and sometimes transport into the anticyclone. Therefore, Taiwan could be located north, south or in the middle of the BB plume. Two events of BB plume transport were addressed. Both events showed that OM accounted for the highest proportion in PM$_{2.5}$ among all of the species. The proportion of EC in PM$_{2.5}$ and NH$_4^+$ in PM$_{2.5}$ increased near the fire source and then remained nearly constant during transport. Even when the BB parcel was near fire sources, NO$_x$ was converted into HNO$_3$, NO$_3^-$, PAN, and PANX. Along the long range transport, nitrate decreased and relatively sulfate increased. Meanwhile, air from Asia also influenced SO$_4^{2-}$.

On the other hand, since the carbonaceous content dominates the BB aerosols, the organic chemistry in air quality models like CMAQ is needed to be improved in order to illustrate the SOA production. In addition, the FLAMBE BB emission inventory was reduced by a factor of 4 in current study. The sensitivity test suggested the deviation of emission inventory should be resolved for future studies.

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