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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.

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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 $PM_{2.5}$, consistent with the observations at both near-source regions and Mt. Lulin. For nitrogen species, nearly all NO_x molecules oxidized into HNO_3, NO_3^-, PAN, and PANX in fires or near fires. The synchronic consumption of NO_x, SO_2, and NH_3 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 $PM_{2.5}$ and elemental carbon to $PM_{2.5}$ remained nearly constant because additional sources were lacking.

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

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earth (Jacobson, 2001; Gadhavi and Jayaraman, 2010). BB may also cause surface cooling that is related to direct aerosol effects (Sakaeda et al., 2011). Therefore, research on BB pollutants has been gaining attention in recent years. For example, Sciare et al. (2008) observed organic carbon (OC) and BC in Eastern Mediterranean from 2001 to 2006 and found BB aerosols from agricultural wastes burning in European countries around the Black Sea that could travel to Crete Island. Arola et al. (2007) used MODIS satellite data and HYSPLIT model (Draxler and Rolph, 2013) to study the transport of BB aerosols from Eastern Europe to Northern Europe, and the exerted considerable effect on surface radiation. In recent years, the BB around Southeast Asia (SEA) and South Asia was studied. The Seven Southeast Asian Studies (7-SEAS) Mission (http://7-seas.gsfc.nasa.gov/) began in 2010 to study the effect of BB aerosols.

Although Lin et al. (2013), Reid et al. (2013), and other articles published in *Atmospheric Environment* volume 78 and *Atmospheric Research* volume 122 have addressed the properties of BB pollutants in SEA, these references only provide the chemical, physical, optical, and radiative characteristics of BB aerosols and related pollutants in specific sources and downwind locations. Although few studies have used chemical transport models to explain the chemical field of BB pollutants, Fu et al. (2012) applied the CMAQ model to assess the effect of BB aerosols from SEA on East Asia (EA). Their simulations showed that the percentage of effect via long-range transport on downwind regions, including the Pearl River Delta region and Fujian province, could reach 20%–50% on CO, 10%–30% on O_3, and as high as 70% on $PM_{2.5}$. Huang et al. (2013) explained the same event further and suggested that BB in SEA would affect a large area through long-range transport. The contribution of BB to AOD in the downwind regions was significant and ranged from 26% to 62%. However, the chemical evolution of BB aerosol and related chemical species during long-range transport has not been resolved to date.

With regard to the transport of BB pollutants, Lin et al. (2013) implemented the 32-year (1979–2010) monthly mean atmospheric circulation with the National Centers for Environmental Prediction-II reanalysis at 2.5° longitude × 2.5° latitude winds (Kanamitsu et al., 2002) and found that the streamlines that indicated southerly winds at 925 and 850 hPa over Indochina would couple during March and April. The southerly winds climbed over the mountains in northern Indochina and became an upslope wind, which benefits the rise of BB pollutants to approximately 700 hPa. Lin et al. (2009) further suggested that BB pollutants near the surface in Indochina could be driven using the upward transport in the leeside trough on the east side of Tibet Plateau below 3 km and by the horizontal transport in the strong westerly winds prevailing above 3 km. Most of the previous studies focused on the rising mechanism of transporting BB pollutants from the surface to the atmosphere (Lin et al., 2009, 2013; Yen et al., 2013). Recently, Fu et al. (2012) applied the CMAQ model to analyze the transport pathways of BB aerosols from SEA to EA during the 2006 BASE-ASIA campaign (Biomass Burning Aerosols in Southeast Asia: Smoke Impact Assessment) of NASA. Fu et al. (2012) found a slight difference on the effect of BB on the Yangtze River Delta region between March 2006 and April 2006. In March 2006, the effect of BB was mainly concentrated in SEA and southern China. In April 2006, the westerly winds flowing at low latitudes became southwesterly winds that flowed above 15° N, which could push the SEA outflows northward.

In this study, we use data from the 2010 Dongsha experiment (Wang et al., 2011; Lin et al., 2013), which is the preliminary study of the 7-SEAS mission. We first re-analyzed the horizontal transport of BB pollutants from SEA to downwind regions in high altitudes. We then explained the chemical evolution of BB pollutants in BB 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 resolutions 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

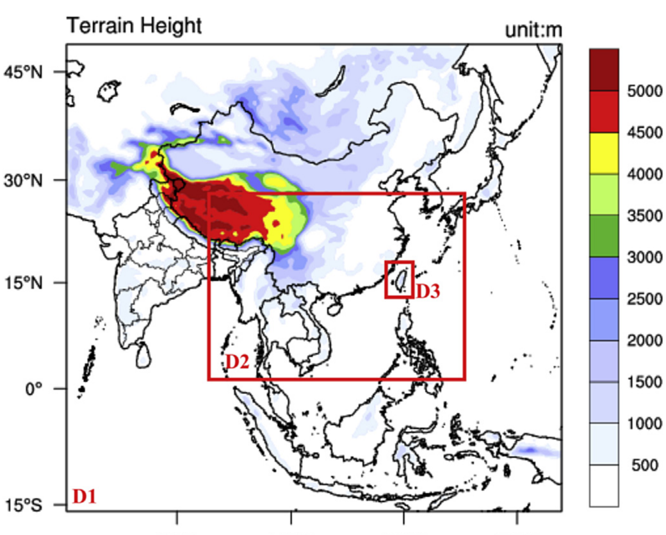

Fig. 1. Three nested domains for current simulation.