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Influence of anthropogenic aerosols on the Asian monsoon: a case study using the WRF-Chem model

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Abstract

Aerosols, in particular those related to anthropogenic activities, including black carbon, organic carbon, and sulfate aerosols, have been found to affect the Asian monsoon through direct and indirect aerosol radiative forcing. In this work, we use the coupled regional Weather Research and Forecasting model with Chemistry (WRF-Chem) to understand how aerosol changes from local emission sources could modulate the Asian monsoon precipitation through aerosol direct and indirect radiative effects. Our modeling results with the consideration of the local emissions show an improvement in simulated monsoon precipitation, when compared to reanalysis data and satellite observations. Aerosols generally induce a reduction in pre-monsoon and monsoon precipitation in East Asia. Over the Indian region, local anthropogenic emissions tend to reduce precipitation in the source regions while slightly increasing precipitation outside of the emission source regions. The increase in precipitation corresponds to a decrease in the cloud base level or lifting condensation level. Analysis of vertical cloud properties suggests that the increased cloud droplet number and prolonged cloud lifetime/reduced precipitation efficiency due to the local aerosol emissions are responsible for the precipitation reduction over East Asia. Aerosols from local emissions also play a very important role in the simulated surface temperature, radiation, and monsoon circulations.

1 Introduction

Aerosols directly affect the atmospheric radiation budget because they scatter and absorb solar radiation. In addition, aerosols, acting as cloud condensation nuclei, can alter the number and size of cloud particles, modifying the albedo of clouds and potential precipitation. Aerosols represent a major source of uncertainty in understanding past climate change and projecting future climate (IPCC, 2007). Recent studies have shown that aerosols can cause substantial modification of the energy balance at the top of the

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atmosphere and the Earth's surface, thus changing cloudiness and the hydrologic cycle (Ramanathan et al., 2001; Breon, 2006; Li et al., 2011). Aerosols from pollution tend to increase the number of small cloud droplets and enhance cloud lifetime as they are less efficiently converted to raindrops (Albrecht, 1989; Pincus and Baker, 1994; 5 Haywood and Boucher, 2000), which is often called the second indirect effect. Satellite observations show that there is a positive and robust relation between aerosol loading and cloud cover (Kaufman and Koren, 2006). Through the indirect effects, aerosols have been shown to affect precipitation frequency and rain rates by exerting radiative forcing and changing cloud properties (e.g., Levi and Cotton, 2009; Li et al., 2012; Tao et al., 2012).

The Asian monsoon system, which provides important water resources in Asia, extends over subtropics and mid-latitudes. Observations have suggested that the Asian summer monsoon circulation and precipitation have declined since the 1970s (Ding et al., 2007). The cause of this decline may be associated with aerosols, thus the 15 effects of aerosols on the Asian monsoon system have been the major topic of extensive research recently (e.g., Ramanathan et al., 2001; Lau and Kim, 2006; Lau, 2008; Lelieveld et al., 2001; Randles and Ramaswamy, 2008; Meehl et al., 2008; Kuhlmann and Quaas, 2010; Li et al., 2011; Ganguly et al., 2012). For example, using a global climate model (GCM), Lau and Kim (2006) pointed out that anthropogenic aerosols combined with natural dust aerosols accentuate the elevated heating, enhance the 20 meridional temperature gradient, and contribute to increased rainfall over India during the pre-monsoon (defined as March, April, and May, MAM) and summer monsoon (defined as June, July, and August, JJA) seasons. Meehl et al. (2008) showed that absorbing aerosols like black carbon have likely contributed to the observed decreasing 25 precipitation trends over parts of the Indian Asian monsoon region. Wang et al. (2009) found that absorbing anthropogenic aerosols resulted in a northward shift of modeled convective precipitation through their influence on the moist static energy in the sub-cloud layer. Collier and Zhang (2009) found that the inclusion of aerosols in a global climate model (CAM3) results in drops in surface temperature and increases in pre-

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cipitation over central India during the pre-monsoon season. The presence of aerosols induces tropospheric shortwave heating over central India, which destabilizes the atmosphere for enhanced convection and precipitation. Zhang et al. (2011) used the 5 same model and found that aerosols tend to reduce precipitation in India and China through direct radiative effects. Based on global climate model simulations, these studies have shown that aerosols can exert different influences on precipitation through direct and absorbing effects; however, they did not explore the indirect aerosol effects on the Asian monsoon system.

Because of lower confidence in our understanding of the indirect effects of aerosols 10 (Levi and Cotton, 2009), the indirect effects and possible interactions with the monsoon system remain largely unknown. Recently, a few studies (e.g., Chen et al., 2010; Cowan and Cai, 2011; Bollasina et al., 2011; Ganguly et al., 2011) have started using global models to examine the indirect effects of aerosols on the monsoon system. Using a global climate model with direct and indirect aerosol feedback, Bollasina 15 et al. (2011) found a weakening of the South Asian summer monsoon and an associated decrease in precipitation in areas including Northern India and Southern China. They attributed this to human-influence aerosol emissions through a slowdown of the tropical meridional circulation. By running the latest version of CAM5 with aerosol direct and indirect effects, Ganguly et al. (2012) found that anthropogenic aerosols from 20 the local emissions within Asia contribute to the overall reduction in the mean summer monsoon precipitation in the South Asian monsoon region. Using a high-resolution ($0.83^\circ \times 1.25^\circ$) global environment model, Guo et al. (2013) studied the effect of regional changes in anthropogenic aerosols on East Asian monsoon precipitation and found that changes in sulfate and BC aerosols resulted in different responses in surface 25 temperature and precipitation when considering direct and indirect aerosol forcings. These studies considering aerosol indirect effects suggest an overall reduction in the mean summer monsoon precipitation in the Asian monsoon system, but different types of aerosols tend to have different impacts.

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Clearly, there is inconsistency in the results from previous studies on the impacts of aerosols on the Asian monsoon system. The net impacts of aerosols on clouds and precipitation have not yet been identified. Accurate estimate of aerosol effects on climate require both aerosol distributions and their interactions with climate. The interactions between aerosols and climate depend on the vertical distribution of aerosols. The lack of detailed measurement of aerosol vertical distribution over large scales also limits our understanding of the roles of aerosols in climate. Most above-mentioned modeling studies about the impacts of aerosols on the Asian monsoon system used prescribed aerosol emissions from offline calculation without the interactions between emissions/aerosols and atmospheric dynamics. The impact of climate change on aerosols is not considered. Also, these studies used GCMs, which have relatively lower spatial resolutions ($\sim 2^\circ \times 2^\circ$) and have difficulty capturing subtle characteristics in the areas of complex terrain, the land use/land cover types, and regional high aerosol concentrations in great details (e.g., Ganguly et al., 2012). The observed maximum rainfalls during the monsoon season along the west coast of India, the North Bay of Bengal, and Northeast India are poorly simulated by most GCMs (Christensen et al., 2007; Kripalani et al., 2007; Ganguly et al., 2012). Kumar et al. (2006) suggested that the difficulty of GCMs in reproducing monsoon precipitation is likely caused by the coarse resolutions of the GCMs, which are not able to correctly represent the regional forcings such as the steep topography of the Himalayas and the Western Ghats.

The advantages of a high-resolution regional model can compensate for the defect of a global model in this regard. Studies have shown that high-resolution regional climate models have better simulation capabilities than the global models for the monsoon climate in Asia (Gao et al., 2008; Zhou and Yu, 2006; Lucas-Picher et al., 2011). In this study, we use a regional online-coupled land-atmosphere-chemistry model to examine the direct and indirect effects of aerosols from local emission sources on Asian monsoon precipitation. The online atmospheric chemistry and calculation of aerosols allow the interactions between aerosols and meteorology (Zhang, 2008).

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The main objective of this work is to understand the role of aerosols emitted from Asia on the Asian monsoon precipitation. The paper is organized as follows. We begin by describing the regional-scale model and its configuration for the simulations. We then evaluate the basic meteorology parameters and aerosol properties against available observations. The impacts of aerosols on precipitation are assessed by examining the radiative forcing, cloud properties, precipitation, and atmospheric circulation.

2 Methodology

2.1 Model description

An online-coupled land-atmosphere-chemistry model, the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) (Grell, et al., 2005; Fast et al., 2006), version 3.3.2, was used in this study. The chemistry and meteorology components of this modeling system employed the same transport, grid, boundary layer, land surface model, and time step. Thus, the continuity equations for all chemical species were performed "online." In this study, we used the mass coordinate version of the model, Advanced Research WRF (ARW).

Gas-phase atmospheric chemistry used in this study is based on the CBM-Z mechanism (Zaveri and Peters, 1999), which includes 67 prognostic species and 164 reactions in a lumped structure approach that classifies organic compounds according to their internal bond types. The fast-J photolysis scheme is used for photolytic reactions within CBM-Z (Wild et al., 2000; Barnard et al., 2004). A sectional approach is used to represent aerosol size distributions using the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC). The aerosol species simulated in MOSAIC include sulfate, nitrate, ammonium, organic carbon (OC), black carbon (BC), and water. Each particle size distribution bin is assumed to be internally mixed, i.e., all particles within a bin are assumed to have the same chemical composition. Both particle mass and number are simulated for each bin. Because bins are based on dry particle diameters, water uptake

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or loss will not transfer particles between bins; however, particle growth or reduction due to chemical reactions, uptake/release of trace gases, or coagulation will produce such transfers. We used four discrete size bins, instead of the 8-bin configuration, with MOSAIC to save computational expense.

5 Aerosol-radiation interactions including direct scattering and absorption, as well as semi-direct effects are implemented in WRF-Chem (Fast et al., 2006). Aerosols affect radiation by changing aerosol optical properties. Each MOSAIC aerosol chemical constituent is linked with aerosol optical properties through a complex refractive index. The refractive index was calculated by volume averaging for each size bin following a refractive index mixing rule described in Bond et al. (2006). The details about the refractive indices used in WRF-Chem are described in Barnard et al. (2010). Space- and time-dependent aerosol optical properties including extinction efficiency, single-scattering albedo, and asymmetry factor at different wavelengths (300, 400, 600, and 1000 nm) are calculated using Mie theory. Wet particle diameters are used in the calculations. 10 Then, the calculated aerosol distributions and associated radiative properties are transferred to the Goddard shortwave radiative transfer model to calculate the direct aerosol effects on radiative forcing (Chou et al., 1998).

Aerosol-cloud interactions are implemented by calculating the activation and resuspension between dry aerosols and cloud droplets (Gustafson et al., 2007; Chapman et al., 2009), which is similar to the method used in the MIRAGE (Model for Integrated Research on Atmospheric Global Exchanges) general circulation model (Ghan et al., 2001). In WRF-Chem, the indirect effects are activated through a prognostic treatment of cloud droplet number. The aerosol particles are activated to form cloud droplets and, aqueous-phase chemistry, and tie a two-moment treatment of cloud water (cloud water mass and cloud droplet number) to precipitation and an existing radiation scheme. 20 Here, the Lin et al. (1983) cloud physics parameterization has been modified to represent both number and mass of cloud water (Chapman et al., 2009). Thus, aerosols affect cloud droplet number and cloud radiative properties (Gustafson et al., 2007; Saide et al., 2011). Clouds also alter aerosol size and composition via aqueous chemistry, wet 25

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scavenging, and gas-phase related photolysis rates. Aqueous chemistry follows the mechanism of Fahey and Pandis (2001) in a bulk approach. There are 50 aqueous-phase species, 17 aqueous-phase ionic equilibria, 21 gas-phase/aqueous-phase reversible reactions, and 109 aqueous-phase chemical reactions. Aerosol particles can transfer between size bins through aqueous chemistry processes. 5

Dry deposition for trace gases uses a surface resistance parameterization developed by Wesely (1989), and for aerosols it is based on Binkowski and Shankar (1995). Wet deposition for both trace gases and aerosols includes both in-cloud and below-cloud wet removal processes. Within cloud, the cloud-borne aerosols and the fraction of trace gases dissolved in cloud water are collected by rain, graupel, and snow, using the corresponding first order loss rate of cloud water from the Lin microphysics scheme. Below-cloud scavenging of aerosols by impaction/interception and selected trace gases by mass transfer have been implemented using the approach of Easter et al. (2004). All trace gas and aerosol species that are scavenged by precipitation are assumed to be immediately wet-deposited and removed from the model. 10 15

2.2 Model configuration

In this study, WRF-Chem is configured to cover the Asian monsoon region including South and East Asia (Fig. 1) with 176×126 grid points at a 42 km horizontal spatial resolution. The model was run from 27 February to 31 August 2008 covering the pre-monsoon and monsoon periods, and the first two days in February were used to spin up the chemical species following the approach used in Jiang et al. (2008). Chemical boundary conditions were obtained from 6 hourly MOZART-4 global simulations (Emmons et al., 2010). The modified Lin et al. (1983), which includes a prognostic treatment of cloud droplet number (Ghan et al., 1997), is used to simulate the aerosol indirect effect. The Lin microphysics scheme and the Goddard short wave radiation scheme (Chou et al., 1998; Fast et al., 2006; Chapman et al., 2009) were used to support aerosol direct, indirect, and semi-direct feedbacks to meteorology. Other selected parameterizations used are summarized in Table 1, including the Grell-Devenyi 20 25

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Cumulus Parameterization scheme (Grell and Devenyi, 2002), the Yonsei University Planetary Boundary Layer (PBL) scheme (Hong and Pan, 1996), the Rapid Radiative Transfer Model Longwave Radiation scheme (Mlawer et al., 1997), and the Noah Land Surface Model (Chen et al., 1997). The initial meteorological fields and boundary conditions were derived from NCEP (National Centers for Environmental Prediction) Final Analysis data with a $1^\circ \times 1^\circ$ spatial resolution and six-hour temporal resolution. The NOAA weekly sea surface temperature data (Reynolds et al., 2002) were used to update the oceanic conditions in the model.

Anthropogenic emissions of NO_x , SO_2 , VOCs, $\text{PM}_{2.5}$ and PM_{10} for Asia were taken from the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) inventory (Zhang et al., 2009). The 2000 database of Reanalysis of Tropospheric Chemical Composition (RETRO) (<http://retro.enes.org/index.shtml>) at 0.5° was used where INTEX-B inventory data were not available. As anthropogenic emissions data are not available for the study year, 2008, this could give rise to some uncertainty in the model results. The seasonal and diurnal variability in anthropogenic emissions, which has been suggested to play an important role in atmospheric chemistry (e.g., Han et al., 2009), was also not included. This could give rise to uncertainty in the simulated aerosol impacts. Because of the strong seasonal variation in biomass burning activities, the monthly fire emissions were estimated for the simulation period using the Fire Inventory from NCAR (FINN) version 1 (Wiedinmyer et al., 2011). We used the same method in Jiang et al. (2012) to speciate biomass burning emissions to the CBMZ mechanism. Biogenic emissions were calculated online in WRF-Chem using the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.04 (Guenther et al., 2006) and updated maps of isoprene emission factor, leaf area index, and plant functional types from MEGAN 2.1 (Guenther et al., 2012). Other aerosol emissions of dust and sea-salt were not considered, because the main focus of this study is the role of anthropogenic related emissions. Figure 1 shows primary aerosol emissions over the modeling domain. High concentrations of $\text{PM}_{2.5}$ emissions including BC, OC, and SO_4 are seen

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in North India and East China. The high BC and OC emissions in the pre-monsoon season over South East Asia are due to local human-induced biomass burning.

To test the effects of aerosols on the Asian monsoon through aerosol-cloud-precipitation interactions, we performed two simulations. In the control (CTRL) run, local point source emissions including anthropogenic and biomass burning emissions were used to simulate aerosol impacts on the monsoon system. In the sensitivity (EXP) run, the local point source emissions including anthropogenic and fire emissions were turned off or removed. The designed experiments allow us to better identify the key mechanisms and processes involved in the aerosol-cloud-radiation feedbacks. The differences between CTRL and EXP are used to reveal the contribution of local emissions to changes in the monsoon system. One additional experiment included the influence of dust aerosols. As the major focus of this work is to examine the impacts from aerosols related to anthropogenic activities, we only briefly compare the results with the impacts due to dust aerosols which could also have important effects on precipitation as suggested by other studies, particularly in Southeast Asia (e.g., Lau and Kim, 2006).

3 Comparison of baseline simulation with observations

3.1 Aerosol

Several studies (e.g., Fast et al., 2006; Zhao et al., 2011; Jiang et al., 2010, 2012), through comparisons with available satellite and ground-based aerosol measurements, have demonstrated the capability of WRF-Chem to simulate aerosols and their optical properties. Here, we briefly evaluate the model performance in simulating aerosol optical depth (AOD). Figure 2 shows the simulated and satellite (MISR and MODIS) derived AOD in the pre-monsoon and monsoon seasons. MODIS retrieved much higher values of AOD over the regions like Northeast China and North India in the monsoon season. Tripathi et al. (2005) compared MODIS-derived AOD with ground-based AOD measurements and found that MODIS overestimates AOD during the monsoon season

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over the northern part of India. Thus, the high AOD values seen in MODIS during the monsoon season over North India could be related to the biases associated with the MODIS aerosol product. In general, the model captures high AOD values over the high emissions source regions in South and East Asia. The WRF-Chem model tends to underestimate AOD in regions like Northwest Asia owing to the lack of dust emissions from the deserts in this region and anthropogenic emissions from Europe.

The Aerosol Robotic Network (AERONET, Holben et al., 1998) has several ground-based sites in the modeling domain measuring aerosol optical properties (Dubovik et al., 2002). Figure 3 shows the time varying AODs simulated by the model and measured at eight AERONET sites (shown in Fig. 2a). In general, the model captures the AERONET measured AOD, though the model fails to capture the observed high peaks of AOD at some time periods. While inclusion of dust increases the modeled AOD, dust emissions (using the default method in the code) are not enough to give good agreement with the observations. The biases between model results and observations could arise for several reasons. For example, the local anthropogenic emissions used in this study were not for 2008; instead they were based on means of previous years. There is no SOA formation in the aerosol scheme used in this study. Further, there is no diurnal and monthly variability in the anthropogenic emissions. It is likely that BC emissions are underestimated in general. Emissions, such as trash burning (Christian et al., 2010), are not included in current simulations.

3.2 Climate

Figure 4 compares model simulated surface air temperature from the CTRL simulation with ERA_Interim reanalysis data (Dee et al., 2011). The model, when considering both direct and indirect aerosol effects, captures the pattern and magnitude of surface air temperature in comparison to the ERA_Interim reanalysis data. This indicates that if the radiative forcing of local aerosols was not considered, the model would tend to overestimate temperatures in some regions like South and East Asia. Lucas-Picher et al. (2011) found that regional climate models without aerosol feedbacks overestimate sur-

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face air temperature over much of the South Indian monsoon region. It should be noted, though, the model still overestimates the surface air temperature in Northwest India, which is strongly influenced by the nearby Himalayas and the Thar Desert (Ji et al., 2011). The overestimation in temperature is mainly caused by the underestimation of aerosols, which if improved may reduce the temperature bias here.

The Asian monsoon is accompanied by a seasonal change in the direction of the prevailing wind. The wind shift often brings about a marked change in precipitation. Figure 5 shows, for March, May, and July, precipitation from the TRMM (Tropical Rainfall Measuring Mission) satellite measurement and 850 hPa wind circulation from the ERA_Interim reanalysis data. The overall spatial distributions of precipitation in different months are fairly well simulated by the model with aerosol forcing. Some differences between satellite retrieved and model simulated precipitation exist in some regions (e.g., Northwest India). This could be due to biases associated with satellite products or the model. More details regarding how aerosols affect precipitation are discussed in Sect. 4.3. The WRF-Chem model does simulate the seasonal changes in wind direction, though the magnitude of wind speed is overestimated in May and July over the southern part of the modeling domain. Several previous studies (e.g., Roux et al., 2009; Mass and Ovens, 2010, 2011; Mölders et al., 2012) also found that WRF or WRF-Chem generally overestimates wind speed in flat areas.

4 Model Results

4.1 Radiative forcing of anthropogenic aerosols

Figure 6 shows simulated differences in downward shortwave radiation reaching the surface, outgoing longwave radiation (OLR), and surface air temperature (or 2 m air temperature) in the pre-monsoon and monsoon seasons between the CTRL and EXP simulations. In this particular year (2008), aerosols from anthropogenic and fire sources reduce the downward solar radiation reaching the surface by up to 80 W m^{-2} .

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The downward solar radiation reaching the surface plays an important role in affecting the near-surface air temperature. Significant cooling (1–3 K) near the surface is widespread, and is seen over the areas where strong aerosol emissions and reduced downward solar radiation occur. However, the cooling is not necessarily restricted to the emission source regions (e.g. the Tibetan Plateau during the monsoon season). Fast and non-uniform responses resulting from aerosol forcing could also affect temperature changes in clouds regionally (Jiang et al., 2012).

The spatial distributions of changes in radiation and surface temperature due to local aerosols are different for the pre-monsoon and monsoon seasons. During the pre-monsoon season, reductions in radiation and temperature are more pronounced over the emission source regions like Southeast China and Southeast Asia. During the monsoon season, the reduction in downward solar radiation over India is enhanced, and so is surface air temperature. We also see cooling outside of the emission source regions. These differences in changes in radiation and temperature are somewhat attributed to different aerosol loadings in the different seasons as a result of the circulation shifts associated with the monsoon system. Because of the aerosol indirect effect, changes in cloud properties further complicate the changes in surface air temperature. The smaller decrease in surface air temperature in the monsoon season in regions like Southeast Asia is related to an increase in monsoon precipitation and associated reduction in aerosol concentrations as compared to the pre-monsoon season. Ganguly et al. (2012) also found that aerosols from Asia lead to significant and widespread surface cooling and the cooling areas are over areas with high AOD and clouds. They also showed that there was a strong correlation between changes in aerosols and downward solar radiation reaching the surface, and the relationship between aerosol change and temperature was more non-linear. The feedback processes involved complicate the relationships.

OLR (Fig. 6b and e), which is strongly related to precipitation (Ding and Chan, 2005), is also affected by the local anthropogenic aerosol emissions and increases over East Asia for both the pre-monsoon and monsoon seasons. In the monsoon season, the

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increase in OLR is larger compared to the pre-monsoon season, suggesting the increased response in precipitation due to aerosol changes. Over the Indian region, in the regions outside of the high emission source regions, OLR is decreased due to the presence of aerosols emitted within the domain, suggesting an increase in precipitation. The change in precipitation and its relationships with OLR will be further discussed in Sect. 4.3.

4.2 Local anthropogenic aerosol impacts on clouds

Not only do aerosols affect cloud characteristics via direct radiative effects, but they also affect clouds by altering the number of cloud condensation nuclei that cloud drops form on (Twomey, 1977; IPCC, 2007). To better understand the changes in precipitation caused by changing aerosol loading, detailed analysis of cloud properties over different regions is presented. Figure 7 shows vertical cross-section plots of the changes in cloud drop number concentrations, cloud water mixing ratio, rainwater mixing ratio, and upward motion over the highlighted regions in India and East Asia (IN and EA) (shown in Fig. 6a). In the pre-monsoon season, over IN, aerosols from local sources are restricted to the near surface layers, resulting in a small increase in cloud droplet number and cloud water mixing ratio at lower levels. This could be in part attributed to the weak large-scale circulation in the pre-monsoon season (Fig. 5) and the weak upward motion (reflected by negative W in Fig. 7). The small changes in cloud droplet number concentrations and cloud water mixing ratio along with the decreased upward motion do not lead to changes in rainwater mixing ratio in the India region, suggesting small changes in precipitation. In contrast, over EA, aerosols are lifted to upper layers (up to ~ 600 hPa) to increase cloud drop numbers and cloud water mixing ratios. Because of the second indirect effect of aerosol (Albrecht, 1989), that is larger amount of aerosols lead to smaller cloud droplets and reduce the precipitation efficiency of clouds (Chapman et al., 2009; Haywood and Boucher, 2000), aerosols in this region reduce the rainwater mixing ratio and thus result in less precipitation. The weak upward motion over the EA region also inhibits the formation of precipitation.

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During the monsoon season, aerosols from local sources increase cloud droplet number concentrations and cloud water mixing ratio over both regions (IN and EA). Over IN, the changes in rain mixing ratio due to aerosols are mixed, with some regions experiencing more rainfall while other regions having less rainfall. The changes in rainwater mixing ratio are strongly related to upward motion. The decrease in upward motion due to local aerosol emissions is associated with decreased rainwater mixing ratios or precipitation. The strong increases in cloud droplet number concentrations and cloud water mixing ratio, coupled with reduced amounts of rain mixing ratio suggests that local anthropogenic aerosols are reducing the precipitation efficiency (Lohmann and Feichter, 2005). However, regions that have modest increases in cloud droplet number concentrations and cloud water mixing ratio (18–20° N over IN; 36–38° N over EA) result in more rainfall. This is likely connected to the circulation because upward wind increases in these locations. In EA, the reduction in precipitation and the increase in water vapor (not shown) reduce the precipitation and increase cloud lifetime by reducing precipitation efficiency of cloud through increased small-size cloud drop number concentrations, cloud water mixing ratio (Fig. 7), and water vapor (Lohmann and Feichter, 2005).

4.3 Local anthropogenic aerosol impacts on precipitation

Figure 8 shows, for the pre-monsoon and monsoon seasons, the spatial distributions of precipitation from satellite estimation (Precipitation Estimation from Remotely Sensed Information using Artificial Neural Networks, PERSIANN) and measurements (Tropical Rainfall Measuring Mission, TRMM, Huffman, et al., 2007) observations, and the WRF-Chem simulations. The selection of PERSIANN and TRMM is based on their relatively high spatial resolution and good coverage in the monsoon region. Overall, the spatial distributions of observed precipitation from PERSIANN and TRMM are very similar over much of the modeling domain, but differ in regional details (e.g., high precipitation near the Tibet Plateau at 30° N, 95° E and Northwest IN occurs only in the TRMM observation, which might be related with high bias caused by cloud contamination in

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the TRMM data). The PERSIANN data set is believed to be better than the TRMM data set here due to the fact that the PERSIANN data set has a higher spatial resolution and it uses information derived from different satellite measurements including TRMM.

Over IN, the modeled precipitation responses to aerosol changes are different in different seasons. The simulations with (CTRL) and without (EXP) aerosols from local sources show a similar performance during the pre-monsoon season. During the monsoon season, the aerosols mainly decrease precipitation over the source regions, while slightly increase precipitation outside of the local emission source regions. Over EA, the CTRL experiment produces less precipitation in both pre-monsoon and monsoon seasons, and the simulated precipitation pattern is much closer to the TRMM observation. The response of precipitation to local aerosol emissions through direct and indirect aerosol forcing is consistent with several recent findings (e.g., Chapman et al., 2009; Ganguly et al., 2012). Using a global climate model (CAM5) with direct and indirect effects of aerosols, Ganguly et al. (2012) also found a reduction in mean summer monsoon precipitation over much of the South Asian monsoon region as a result of aerosol forcing from local anthropogenic and biomass burning emissions.

The influence of dust aerosols from dust sources in the west of India and Northern China on precipitation is shown in Fig. 9. Overall, the impacts due to dust aerosols are much smaller as compared to the effects from anthropogenic aerosols. There is a small increase in precipitation in the Indian region, and the changes in East Asia are different in different seasons. While previous studies (e.g., Lau and Kim, 2006) have shown that dust aerosols when combined with other natural aerosols tended to increase precipitation in pre-monsoon and monsoon seasons in India, our simulations do not show significant impacts from dust aerosols. It is possible that the WRF-Chem dust aerosol scheme, which has not been well tested over the Asian region, underestimates dust emissions. Saide et al. (2012) also stated that the default online wind-blown dust model has large biases in simulating dust emissions and AOD and poor model representation of dust compositions, which are important for the aerosol indirect effects. In future stud-

ies, when the better dust scheme is available, it is important to include and compare the dust aerosol impacts.

The latitude-time plot of precipitation (Fig. 10a and b), which shows the difference of zonally averaged precipitation, clearly shows that local aerosols tend to increase monsoon season precipitation over the regions outside of the emission source locations in IN. Near the emission source region ($\sim 24^\circ$ N), precipitation decreases in response to the increase in aerosols. Similarly, Wang et al. (2009) found that absorbing aerosols created a stronger convective precipitation in the Northern Indian region and a significant reduction in southern India, suggesting different responses in precipitation to aerosol changes. Over much of EA, local aerosols reduce precipitation throughout the pre-monsoon and monsoon seasons. This is consistent with the spatial patterns of precipitation changes. Daily averages of OLR over the two regions are used as a proxy for large-scale convective activity (Ding and Chan, 2005). Figure 10c and d shows that the changes in the precipitation evolution strongly correlate with the evolution of OLR changes in the two regions, with higher OLR corresponding to lower precipitation. This suggests that when precipitation decreases due to local aerosols, large-scale convective activities are reduced.

As aforementioned, PERSIANN data use information from several satellite measurements and are at higher spatial and temporal resolutions, a comparison of the modeled and PERSIANN measured daily accumulative precipitation over IN and EA is shown in Fig. 11. In general, the temporal development of precipitation is reproduced fairly well in the two experiments at the two regions. The simulation with the local emissions included is in better agreement with the observed trend. Over IN, EXP and CTRL have similar accumulated precipitation in the pre-monsoon season, while they differ significantly from each other starting in July. The result from CTRL is much closer to the observation in the monsoon season. The improvement due to aerosol forcing is also significant over EA. The CTRL simulation produces less precipitation and agrees well with the observation. The EXP run without the influence of local emissions over-

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estimates precipitation in this region. This highlights the role of aerosols in reducing precipitation over EA in both pre-monsoon and monsoon seasons.

Precipitation is strongly influenced by clouds, which is tightly linked to the lifting condensation level (LCL) (Betts, 2007; Jiang et al., 2009) and convective available potential energy (CAPE). LCL, which is the level at which a raising air-parcel becomes saturated, can be used as an indicator of the estimated mean height of cloud base and can help us to evaluate atmospheric stability. Figure 11c shows that, over EA, aerosols not only reduce precipitation but also increase LCL or cloud base height. The increase in LCL is accompanied by the reduction in the amount of clouds that form precipitation. Over IN, aerosols decrease LCL over much of the area except the emission source regions, resulting in an increase in region-averaged precipitation. The sounding plots (Fig. 12) in August 2008 at two representative locations (sites A and B shown in Fig. 6a) in IN and EA also suggest that aerosols reduce precipitation by reducing CAPE from 1100 to 498 (J) at Site B over the EA region and increase precipitation slightly by increasing CAPE from 758 to 927 (J) at Site A over the IN region. The changes in CAPE at the two regions suggest that due to the aerosol effects, convective activities in the monsoon region are altered with reduced activities found over EA and slight increased activities seen outside of the emission source regions in IN.

4.4 Local anthropogenic impacts on atmospheric circulation

Figure 13 shows the changes in the mean spatial distribution of sea level pressure (SLP) and wind vector at 850 hPa due to aerosol emissions in the pre-monsoon and monsoon seasons. In the pre-monsoon season, increases in SLP occur over Southeast and East Asia where the surface cooling caused by aerosols is observed. The cooling (Fig. 6) and the associated increase in SLP lead to an anomalous anticyclonic circulation at 850 hPa over East Asia. The change in circulation reduces the moisture that is being transported by the southwest flow (Fig. 5), resulting in reduced precipitation in this region (Fig. 8). During the monsoon season, the increase in SLP expands to a larger area including the North and Northwest India, and the magnitude of the in-

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crease also increases. Due to this increase in SLP, circulation shifts to flow more from the land to the nearby oceans with local aerosols, leading to less moist air and less precipitation in South, Southeast, North, and Northwest India. This suggests that heating/cooling caused by aerosols from local sources tends to increase the near surface pressure, dry the air, and reduce precipitation.

5 Discussion and conclusions

This study examines the impacts of direct and indirect effects of aerosols on the Asian monsoon precipitation in 2008 using the WRF-Chem model. The model performance is evaluated against satellite measurements and reanalysis data. Generally, the model simulates the spatial pattern of AOD fairly well. The underestimation in regions like Northwest India is related to the lack of emission sources. When considering aerosol-cloud-precipitation feedbacks, the WRF-Chem model is capable of reproducing near surface temperature, precipitation, and monsoon circulations compared to the satellite and reanalysis data.

Through the comparison between the runs with and without local aerosol emission sources, the impacts of aerosols on radiation, temperature, clouds, precipitation, and atmospheric circulation are discussed. Aerosols from local emission sources reduce the downward solar radiation reaching the surface by up to 80 W m^{-2} and surface temperature by 1–3 K in the pre-monsoon and monsoon seasons. The widespread cooling is strongly correlated with the reduced downward solar radiation, but is not necessarily restricted to the emission source regions. The feedback between aerosols and clouds affects the nonuniform responses in the surface temperature. Changes in aerosols also affect OLR, a strong indicator for large-scale convection and precipitation.

The clouds and precipitation responses to local aerosol radiative forcing differ in EA and IN and different seasons. Over IN, clouds and precipitation are not strongly affected by aerosols in the pre-monsoon season, while there is an increase in clouds and precipitation during the monsoon season except for over the emission source re-

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gions. Aerosols decrease precipitation near the emission source regions in IN. Due to aerosol forcing, precipitation decreases over much of EA in both the pre-monsoon and monsoon seasons. The decrease in precipitation is highly correlated with the increases in small cloud droplets, cloud base height (or LCL), and SLP. This can be explained that aerosols from local emission sources can increase small cloud droplets, which tend to have a longer life time, reducing the amount of clouds that could fall as precipitation (Chapman et al., 2009). The cloud base height is also increased due to the reduced CAPE caused by the increase in aerosol concentrations and small cloud droplets.

Overall, this study highlights the importance of aerosol forcing in the Asian monsoon precipitation. Without the consideration of aerosol-cloud-precipitation feedback in regional climate models, the monsoon precipitation will be overestimated in India or underestimated in East Asia. Lucas-Picher et al. (2011) evaluated four regional climate models that did not consider aerosol feedbacks and found that the performance of the models in simulating the Indian Monsoon precipitation was poor. Some models have a warm bias over much of their study domains, and they attributed this to missing or poor representation of regional processes and feedbacks, with aerosol feedbacks being one of them.

While the results presented in this study demonstrated the importance of aerosol-cloud-precipitation feedback in simulating Asian monsoon precipitation, the results are subject to several uncertainties. Besides the uncertainties associated with the parameterization schemes used in this study, the anthropogenic emissions used in this study did not have diurnal, daily, and monthly variations, and they did not exactly represent the specific study period. Amnuaylojaroen et al. (2013) found there were differences in anthropogenic emissions provided by different emission inventories and those differences could lead to different chemical productions in the model. Thus, it is expected that when using different emission inventories, the results may be slightly different. More work needs to be done to assess the influence of anthropogenic emission inventories on this topic. The indirect aerosol effects are not fully considered, because the effects on convective clouds and precipitation are not implemented in the current WRF-

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Chem model. In order to take into account the aerosol effects on convective clouds, the model needs to be run at grid spacing of < 5 km (Morrison and Grabowski, 2011; Fan et al., 2012), which is not feasible for large-scale, long-term climate simulations due to the limitation of computational cost. Simulations of multiple years or ensemble simulation approaches are needed to further address the roles of aerosols in the Asian monsoon system.

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References

- Albrecht, B. A.: Aerosols, cloud microphysics, and fractional Cloudiness, *Science*, 245, 1227–30, 1989.
- Amnuaylojaroen, T., Barth, M. C., Emmons, L. K., Carmichael, G. R., Kreasuwun, J., Prasitwatanaseree, S., and Chantara, S.: Effect of different anthropogenic emission inventories on ozone, carbon monoxide, nitrogen oxide and volatile organic compounds in Southeast Asia, in preparation, 2013.
- Barnard, J. C., Chapman, E. G., Fast, J. D., Schmelzer, J. R., Slusser, J. R., and Shetter, R. E.: An evaluation of the FAST-J photolysis algorithm for predicting nitrogen dioxide photolysis rates under clear and cloudy sky conditions, *Atmos. Environ.*, 38, 3393–3403, 2004.
- Barnard, J. C., Fast, J. D., Paredes-Miranda, G., Arnott, W. P., and Laskin, A.: Technical Note: Evaluation of the WRF-Chem “Aerosol Chemical to Aerosol Optical Properties” Module using data from the MILAGRO campaign, *Atmos. Chem. Phys.*, 10, 7325–7340, doi:10.5194/acp-10-7325-2010, 2010.
- Betts, A. K.: Coupling of water vapor convergence, clouds, precipitation, and land-surface processes, *J. Geophys. Res.*, 112, D10108, doi:10.1029/2006JD008191, 2007.
- Binkowski, F. S. and Shankar, U.: The regional particulate matter model 0.1. Model description and preliminary results, *J. Geophys. Res.*, 100, 26191–26209, 1995.

21403

- Bollasina, M. A., Ming, Y., and Ramaswamy, V.: Anthropogenic aerosols and the weakening of the South Asian summer monsoon, *Science*, 334, 502–505, doi:10.1126/science.1204994, 2011.
- Bond, T. C. and Bergstrom, R. W.: Light absorption by carbonaceous particles: an investigative review, *Aerosol Sci. Tech.*, 40, 27–67, 2006.
- Bond, T. C., Habib, G., and Bergstrom, R. W.: Limitations in the enhancement of visible light absorption due to mixing state, *J. Geophys. Res.*, 111, D20211, doi:10.1029/2006JD007315, 2006.
- Breon, F. M.: How do aerosols affect cloudiness and climate?, *Science*, 313, 623–624, 2006.
- Chapman, E. G., Gustafson Jr., W. I., Easter, R. C., Barnard, J. C., Ghan, S. J., Pekour, M. S., and Fast, J. D.: Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources, *Atmos. Chem. Phys.*, 9, 945–964, doi:10.5194/acp-9-945-2009, 2009.
- Chen, F., Janjic, Z., and Mitchell, K.: Impact of atmospheric surface-layer parameterizations in the new land-surface scheme of the NCEP mesoscale Eta model, *Bound.-Lay. Meteorol.*, 85, 391–421, 1997.
- Chen, W. T., Lee, Y. H., Adams, P. J., Nenes, A., and Seinfeld, J. H.: Will black carbon mitigation dampen aerosol indirect forcing?, *Geophys. Res. Lett.*, 37, L09801, doi:10.1029/2010gl042886, 2010.
- Chou, M. D., Suarez, M. J., Ho, C. H., Yan, M. M. H., and Lee, K. T.: Parameterizations for cloud overlapping and shortwave single-scattering properties for use in general circulation and cloud ensemble models, *J. Climate*, 11, 202–214, 1998.
- Christensen, J. H., Hewitson, B., Busuioac, A., Chen, A., Gao, X., Held, I., Jones, R., Kolli, R. K., Kwon, W.-T., Laprise, R., Magaña Rueda, V., Mearns, L., Menéndez, C. G., Räisänen, J., Rinke, A., Sarr, A., and Whetton, P.: Regional climate projections, *Climate Change 2007: The Physical Science Basis*, edited by: Solomon, S. et al., Cambridge University Press, Cambridge, UK and New York, NY, USA, 847–940, 2007.
- Christian, T. J., Yokelson, R. J., Cárdenas, B., Molina, L. T., Engling, G., and Hsu, S.-C.: Trace gas and particle emissions from domestic and industrial biofuel use and garbage burning in central Mexico, *Atmos. Chem. Phys.*, 10, 565–584, doi:10.5194/acp-10-565-2010, 2010.
- Collier, J. C. and Zhang, G. J.: Aerosol direct forcing of the summer Indian monsoon as simulated by the NCAR CAM3, *Clim. Dynam.*, 32, 313–332, doi:10.1007/s00382-008-0464-9, 2009.

21404

- Cowan, T. and Cai, W.: The impact of Asian and non-Asian anthropogenic aerosols on 20th century Asian summer monsoon, *Geophys. Res. Lett.*, 38, L11703, doi:10.1029/2011GL047268, 2011.
- Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, I., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N. and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, *Q. J. Roy. Meteor. Soc.*, 137, 553–597, doi:10.1002/qj.828, 2011.
- Ding, Y. H. and Chan, J. C. L.: The East Asian summer monsoon: an overview, *Meteorol. Atmos. Phys.*, 89, 117–142, 2005.
- Ding, Y., Wang, Z., and Sun, Y.: Inter-decadal variation of the summer precipitation in East China and its association with decreasing Asian summer monsoon. Part I: Observed evidences, *Int. J. Climatol.*, 28, 1139–1161, doi:10.1002/joc.1615, 2007.
- Dubovik, O. and King, M. D.: A flexible inversion algorithm for retrieval of aerosol optical properties from sun and sky radiance measurements, *J. Geophys. Res.*, 105, 20673–20696, 2000.
- Dubovik, O., Holben, B., Eck, T. F., Smirnov, A., Kaufman, Y. J., King, M. D., Tanré, D., and Slutsker, I.: Variability of absorption and optical properties of key aerosol types observed in worldwide locations, *J. Atmos. Sci.*, 59, 590–608, 2002.
- Easter, R. C., Ghan, S. J., Zhang, Y., Saylor, R. D., Chapman, E. G., Laulainen, N. S., Abdul-Razzak, H., Leung, L. R., Bian, X., and Zaveri, R. A.: MIRAGE: model description and evaluation of aerosols and trace gases, *J. Geophys. Res.*, 109, D20210, doi:10.1029/2004JD004571, 2004.
- Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geosci. Model Dev.*, 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010.
- Fahey, K. M. and Pandis, S. N.: Optimizing model performance: variable size resolution in cloud chemistry modeling, *Atmos. Environ.*, 35, 4471–4478, 2001.

21405

- Fan, J., Rosenfeld, D., Ding, Y., Leung, L. Y. R., and Li, Z.: Potential aerosol indirect effects on atmospheric circulation and radiative forcing through deep convection, *Geophys. Res. Lett.*, 39, L09806, doi:10.1029/2012GL051851, 2012.
- Fast, J. D., Gustafson, W. I., Easter, R. C., Zaveri, R. A., Barnard, J. C., Chapman, E. G., Grell, G. A., and Peckham, S. E.: Evolution of ozone, particulates, and aerosol direct radiative forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol model, *J. J. Geophys. Res.*, 111, D21305, doi:10.1029/2005JD006721, 2006.
- Ganguly, D., Rasch, P. J., Wang, H., and Yoon, J.-H.: Climate response of the South Asian monsoon system to anthropogenic aerosols, *J. Geophys. Res.*, 117, D13209, doi:10.1029/2012JD017508, 2012.
- Gao, X., Shi, Y., Song, R., Giorgi, F., Wang, Y., and Zhang, D.: Reduction of future monsoon precipitation over China: comparison between a high resolution RCM simulation and the driving GCM, *Meteorol. Atmos. Phys.*, 100, 73–86, 2008.
- Ghan, S. J. and Schwartz, S. E.: Aerosol properties and processes: a path from field and laboratory measurements to global climate models, *B. Am. Meteorol. Soc.*, 88, 1059–1083, 2007.
- Ghan, S. J., Easter, R. C., Hudson, J., and Breon, F.-M.: Evaluation of aerosol indirect radiative forcing in MIRAGE, *J. Geophys. Res.*, 106, 5317–5334, 2001.
- Grell, G. A. and Dvénényi, D.: A generalized approach to parameterizing convection combining ensemble and data assimilation techniques, *Geophys. Res. Lett.*, 29, 1693, doi:10.1029/2002GL015311, 2002.
- Grell, G. A., Peckham, S. E., Schmitz, R., McKeen, S. A., Frostb, G., Skamarock, W. C., and Eder, B.: Fully coupled “online” chemistry within the WRF model, *Atmos. Environ.*, 39, 6957–6975, 2005.
- Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), *Atmos. Chem. Phys.*, 6, 3181–3210, doi:10.5194/acp-6-3181-2006, 2006.
- Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, *Geosci. Model Dev.*, 5, 1471–1492, doi:10.5194/gmd-5-1471-2012, 2012.

21406

- Guo, L., Highwood, E. J., Shaffrey, L. C., and Turner, A. G.: The effect of regional changes in anthropogenic aerosols on rainfall of the East Asian Summer Monsoon, *Atmos. Chem. Phys.*, 13, 1521–1534, doi:10.5194/acp-13-1521-2013, 2013.
- Gustafson Jr., W. I., Chapman, E. G., Ghan, S. J., and Fast, J. D.: Impact on modeled cloud characteristics due to simplified treatment of uniform cloud condensation nuclei during NEAQS 2004, *J. Geophys. Res. Lett.*, 34, L19809, doi:10.1029/2007GL030021, 2007.
- Haywood, J. and Boucher, O.: Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: a review, *Rev. Geophys.*, 38, 513–543, 2000.
- Han, K. M., Song, C. H., Ahn, H. J., Park, R. S., Woo, J. H., Lee, C. K., Richter, A., Burrows, J. P., Kim, J. Y., and Hong, J. H.: Investigation of NO_x emissions and NO_x-related chemistry in East Asia using CMAQ-predicted and GOME-derived NO₂ columns, *Atmos. Chem. Phys.*, 9, 1017–1036, doi:10.5194/acp-9-1017-2009, 2009.
- Huffman, G. J., Adler, R. F., Bolvin, D. T., Gu, G., Nelkin, E. J., Bowman, K. P., Hong, Y., Stocker, E. F., and Wolff, D. B.: The TRMM Multisatellite Precipitation Analysis (TMPA): quasi-global, multiyear, combined-sensor precipitation estimates at fine scales, *J. Hydrometeorol.*, 8, 38–55, 2007.
- Holben, B. N., Eck, T. F., Slutsker, I., Tanré, D., Buis, J. P., Stezer, A., Vermote, E., Reagan, Y., Kaufman, U. J., Nakajima, T., Lavenu, F., Jankowiak, I., and Smirnov, A.: AERONET-A federated instrument network and data archive for aerosol characterization, *Remote Sens. Environ.*, 66, 1–16, 1998.
- Hong, S. Y. and Pan, H. L.: Nonlocal boundary layer vertical diffusion in a Medium-Range Forecast Model, *Mon. Weather Rev.*, 124, 2322–2339, 1996.
- Intergovernmental Panel on Climate Change (IPCC): *Climate Change 2007: The Physical Science Basis*, IPCC Secretariat, WMO, Geneva, 2007.
- Ji, Z., Kang, S., Zhang, D., Zhu, C., Wu, J., and Xu, Y.: Simulation of the anthropogenic aerosols over South Asia and their effects on Indian summer monsoon, *Clim. Dynam.*, 36, 1633–1647, 2011.
- Jiang, X., Wiedinmyer, C., Chen, F., Yang, Z.-L., and Lo, J. C.-F.: Predicted impacts of climate and land use change on surface ozone in the Houston, Texas, area, *J. Geophys. Res.*, 113, D20312, doi:10.1029/2008JD009820, 2008.
- Jiang, X., Yang, Z.-L., Liao, H., and Wiedinmyer, C.: Sensitivity of biogenic secondary organic aerosols to future climate change at regional scales: An online coupled simulation, *Atmos. Environ.*, 44, 4891–4907, doi:10.1016/j.atmosenv.2010.08.032, 2010.

21407

- Jiang, X., Wiedinmyer, C., and Carlton, A.: Aerosols from fires: an examination of the effects on ozone photochemistry in the Western United States, *Environ. Sci. Technol.*, 46, 11878–11886, doi:10.1021/es301541k, 2012.
- Kaufman, Y. J. and Koren, I.: Smoke and pollution aerosol effect on cloud cover, *Science*, 313, 655–658, doi:10.1126/science.1126232, 2006.
- Kripalani, R. H., Oh, J. H., Kulkarni, A., Sabade, S. S., and Chaudhari, H. S.: South Asian summer monsoon precipitation variability: coupled climate model simulations and projections under IPCC AR4, *Theor. Appl. Climatol.*, 90, 133–159, doi:10.1007/s00704-006-0282-0, 2007.
- Kuhlmann, J. and Quaas, J.: How can aerosols affect the Asian summer monsoon? Assessment during three consecutive pre-monsoon seasons from CALIPSO satellite data, *Atmos. Chem. Phys.*, 10, 4673–4688, doi:10.5194/acp-10-4673-2010, 2010.
- Kumar, K. R., Sahai, A. K., Kumar, K. K., Patwardhan, S. K., Mishra, P. K., Revadekar, J. V., Kamala, K., and Pant, G. B.: High-resolution climate change scenarios for India, *Curr. Sci. India*, 90, 334–345, 2006.
- Lau, K.-M. and Kim, K.-M.: Observational relationships between aerosol and Asian monsoon rainfall, and circulation, *Geophys. Res. Lett.*, 33, L21810, doi:10.1029/2006GL027546, 2006.
- Lau, K., Ramanathan, V., Wu, G., Li, Z., Tsay, S., Hsu, C., Sikka, R., Holben, B., Lu, D., Tartari, G., Chin, M., Koudelova, R., Chen, H., Ma, Y., Huang, J., Taniguchi, K., and Zhang, R.: The joint aerosol-monsoon experiment: a new challenge for monsoon climate research, *B. Am. Meteorol. Soc.*, 89, 369–383, doi:10.1175/BAMS-89-3-369, 2008.
- Lelieveld, J., Crutzen, P. J., Ramanathan, V., Andreae, M. O., Brenninkmeijer, C. M., Campos, T., Cass, G. R., Dickerson, R. R., Fischer, H., de Gouw, J. A., Hansel, A., Jefferson, A., Kley, D., de Laat, A. T., Lal, S., Lawrence, M. G., Lobert, J. M., Mayol-Bracero, O. L., Mitra, A. P., Novakov, T., Oltmans, S. J., Prather, K. A., Reiner, T., Rodhe, H., Scheeren, H. A., Sikka, D., and Williams, J.: The Indian Ocean experiment: widespread air pollution from South and Southeast Asia, *Science*, 291, 1031–1036, 2001.
- Levin, Z. and Cotton, W. R.: *Aerosol Pollution Impact on Precipitation*, 386 pp., Springer, New York, doi:10.1007/978-1-4020-8690-8, 2009.
- Li, Z., Niu, F., Fan, J., Liu, Y., Rosenfeld, D., and Ding, Y.: Long-term net impacts of aerosols on cloud and precipitation, *Nat. Geosci.*, 4, 888–894, doi:10.1038/ngeo1313, 2011.
- Lin, Y. L., Farley, R. D., and Orville, H. D.: Bulk parameterization of the snow field in a cloud model, *J. Clim. Appl. Meteorol.*, 1983, 22, 1065–1092, 1983.

21408

- Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, *Atmos. Chem. Phys.*, 5, 715–737, doi:10.5194/acp-5-715-2005, 2005.
- Lucas-Picher, P., Christensen, J. H., Saeed, F., Kumar, P., Asharaf, S., Ahrens, B., Wiltshire, A., Jacob, D., and Hagemann, S.: Can regional climate models represent the Indian monsoon?, *J. Hydrometeorol.*, 12, 849–868, doi:10.1175/2011JHM1327.1, 2011.
- 5 Mass, C. and Ovens, D.: WRF model physics: problems, solutions and a new paradigm for progress, Preprints, WRF Users' Workshop, NCAR, Boulder, CO, 2010.
- Mass, C. and Ovens, D.: Fixing WRF's high speed wind bias: a new subgrid scale drag parameterization and the role of detailed verification, Preprints, 24th Conf. on Weather and Forecasting/20th Conf. on Numerical Weather Prediction, 22–24 January 2011, Seattle, WA, USA, Amer. Meteor. Soc., 9B.6, 2011.
- 10 Meehl, G. A., Arblaster, J. M., and Collins, W. D.: Effects of black carbon aerosols on the Indian monsoon, *J. Climate*, 21, 2869–2882, doi:10.1175/2007JCLI1777.1, 2008.
- Mlawer, E. J., Taubman, S. J., Brown, P. D., Iacono, M. J., and Clough, S. A.: Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave, *J. Geophys. Res.*, 102, 16663–16682, 1997.
- Morrison, H. and Grabowski, W. W.: Cloud-system resolving model simulations of aerosol indirect effects on tropical deep convection and its thermodynamic environment, *Atmos. Chem. Phys.*, 11, 10503–10523, doi:10.5194/acp-11-10503-2011, 2011.
- 20 Pincus, R. and Baker, M. B.: Effect of precipitation on the albedo susceptibility of clouds in the marine boundary layer, *Nature*, 372, 250–252, doi:10.1038/372250a0, 1994.
- Ramanathan, V., Crutzen, P. J., Kiehl, J. T., and Rosenfeld, D.: Aerosols, climate, and the hydrological cycle, *Science*, 294, 2119–2124, 2001.
- Randles, C. A. and Ramaswamy, V.: Absorbing aerosols over Asia: a geophysical fluid dynamics laboratory general circulation model sensitivity study of model response to aerosol optical depth and aerosol absorption, *J. Geophys. Res.*, 113, D21203, doi:10.1029/2008JD010140, 2008.
- 25 Reynolds, R. W., Rayner, N. A., Smith, T. M., Stokes, D. C., and Wang, W.: An improved in situ and satellite SST analysis, *J. Climate*, 15, 1609–1625, 2002.
- 30 Roux, G., Liu, Y., Monache, L. D., Sheu, R.-S., and Warner, T. T.: Verification of high resolution WRF-RTFDDA surface forecasts over mountains and plains, Preprints, WRF Users' Workshop, Boulder, CO, NCAR, 2009.

21409

- Kumar, K. R., Sahai, A. K., Kumar, K. K., Patwardhan, S. K., Mishra, P. K., Revadekar, J. V., Kamala, K., and Pant, G. B.: High-resolution climate change scenarios for India for the 21st century, *Curr. Sci. India*, 90, 334–345, 2006.
- 5 Saide, P. E., Spak, S. N., Carmichael, G. R., Mena-Carrasco, M. A., Yang, Q., Howell, S., Leon, D. C., Snider, J. R., Bandy, A. R., Collett, J. L., Benedict, K. B., de Szoeke, S. P., Hawkins, L. N., Allen, G., Crawford, I., Crosier, J., and Springston, S. R.: Evaluating WRF-Chem aerosol indirect effects in Southeast Pacific marine stratocumulus during VOCALS-REx, *Atmos. Chem. Phys.*, 12, 3045–3064, doi:10.5194/acp-12-3045-2012, 2012.
- Tao, W.-K., Chen, J.-P., Li, Z.-Q., Wang, C., and Zhang, C.-D.: The impact of aerosol on convective cloud and precipitation, *Rev. Geophys.*, 50, RG2001, doi:10.1029/2011RG000369, 2012.
- 10 Tripathi, S. N., Dey, Sagnik, Chandel, A., Srivastava, S., Singh, Ramesh P., and Holben, B. N.: Comparison of MODIS and AERONET derived aerosol optical depth over the Ganga Basin, India, *Ann. Geophys.*, 23, 1093–1101, doi:10.5194/angeo-23-1093-2005, 2005.
- 15 Twomey, S. A.: The influence of pollution on the shortwave albedo of clouds, *J. Atmos. Sci.*, 34, 1149–1152, 1977.
- Wang, B., Ding, Q., Joseph, P. V.: Objective definition of the Indian summer monsoon onset, *J. Climate*, 22, 3303–3316, 2009.
- Wesley, M. L.: Parameterization of surface resistance to gaseous dry deposition in regional numerical models, *Atmos. Environ.*, 23, 1293–1304, 1989.
- 20 Wiedinmyer, C., Akagi, S. K., Yokelson, R. J., Emmons, L. K., Al-Saadi, J. A., Orlando, J. J., and Soja, A. J.: The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning, *Geosci. Model Dev.*, 4, 625–641, doi:10.5194/gmd-4-625-2011, 2011.
- 25 Wild, O., Zhu, X., and Prather, M. J.: Fast-J: Accurate simulation of in- and below-cloud photolysis in tropospheric chemical models, *J. Atmos. Chem.*, 37, 245–282, 2000.
- Zaveri, R. A. and Peters, L. K.: A new lumped structure photochemical mechanism for large-scale applications, *J. Geophys. Res.*, 104, 30387–30415, 1999.
- 30 Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emissions in 2006 for the NASA INTEX-B mission, *Atmos. Chem. Phys.*, 9, 5131–5153, doi:10.5194/acp-9-5131-2009, 2009.

21410

- Zhang, Y.: Online-coupled meteorology and chemistry models: history, current status, and outlook, *Atmos. Chem. Phys.*, 8, 2895–2932, doi:10.5194/acp-8-2895-2008, 2008.
- Zhang, Y. X., Sun, S., Olsen, S. C., Dubey, M. K., and He, J. H.: CCSM3 simulated regional effects of anthropogenic aerosols for two contrasting scenarios: rising asian emissions and global reduction of aerosols, *Int. J. Climatol.*, 31, 95–114, doi:10.1002/joc.2060, 2011.
- 5 Zhao, C., Liu, X., Ruby Leung, L., and Hagos, S.: Radiative impact of mineral dust on monsoon precipitation variability over West Africa, *Atmos. Chem. Phys.*, 11, 1879–1893, doi:10.5194/acp-11-1879-2011, 2011.
- 10 Zhou, T. J. and Yu, R. C.: Twentieth century surface air temperature over China and the globe simulation by coupled climate models, *J. Climate*, 19, 5843–5858, 2006.

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Table 1. Selected WRF-Chem configuration options used in both (CTRL and EXP) simulations.

Process	WRF-Chem Option
Gas-phase chemistry	CBM-Z
Aerosol chemistry	MOSAIC
Photolysis	Fast-J
Cloud microphysics	Lin et al.
Cumulus scheme	Grell–Devenyi ensemble
Land surface model	Noah LSM
Boundary layer scheme	YSU
Surface layer scheme	Monin–Obukov
Longwave radiation	RRTM
Shortwave radiation	Goddard
Aerosol radiative feedbacks	Direct, semi-direct, 1st and 2nd indirect effects
Biogenic emissions	MEGANv2.0.4
Anthropogenic emissions	2000 RETRO and 2006 INTEX-B emissions
Fire emissions	FINNV1
Chemical boundary conditions	MOZART
Initial and boundary meteorological conditions	NCEP FNL (1° × 1°)

21412

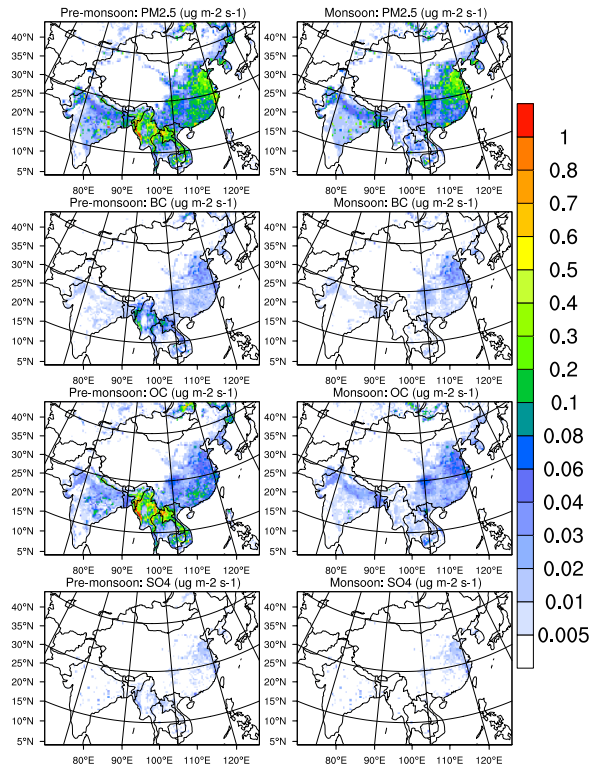


Fig. 1. Emissions of total $PM_{2.5}$, BC, OC, and SO_4 in the pre-monsoon (March, April, and May, MAM) and monsoon (June, July, and August, JJA) seasons in 2008 over the modeling domain.

21413

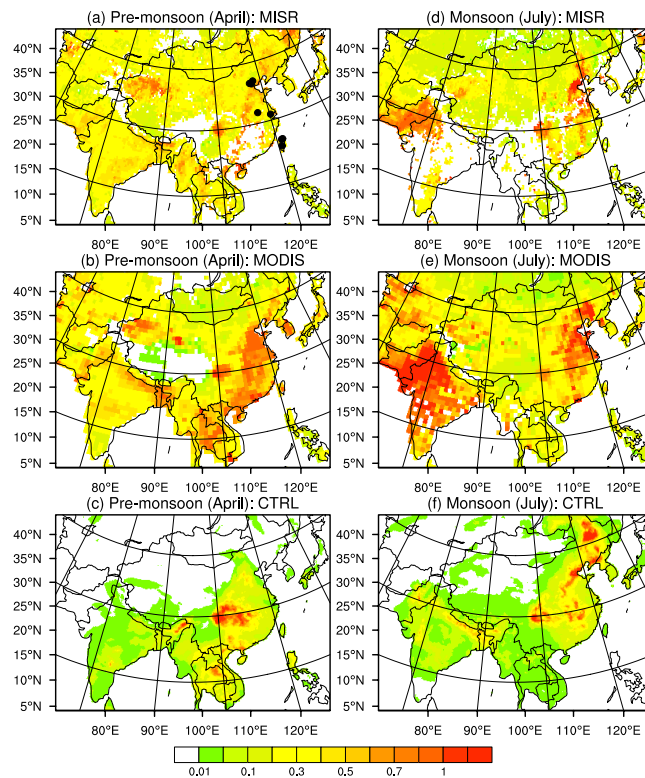


Fig. 2. AODs from the MISR/MODIS satellite measurements (**a**, **d**, **b** and **e**) and the CTRL experiment (**c** and **f**) for selected months (April and July) in pre-monsoon and monsoon seasons in 2008. Black dots on (**a**) are eight AERONET sites.

21414

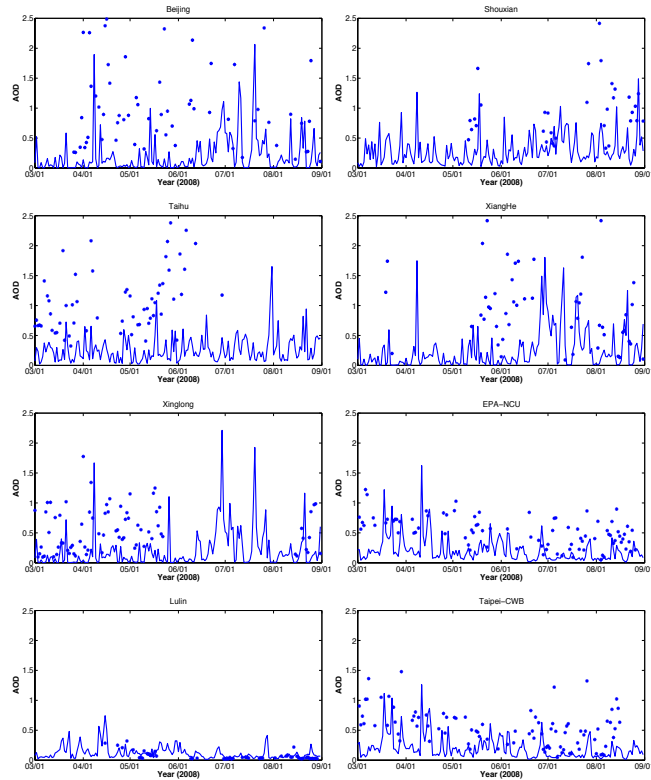


Fig. 3. (a) AODs from CTRL (Solid lines) and AERONET (Dots) at 8 sites (shown in Fig. 2) for the simulation period (May–August 2008).

21415

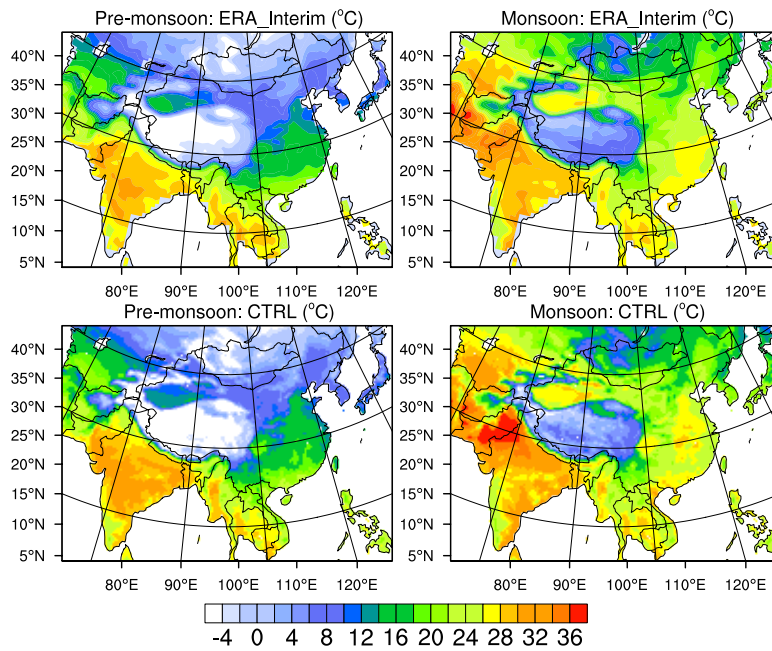


Fig. 4. Surface air temperature (or 2 m temperature) from the ERA-Interim reanalysis and the CTRL in the pre-monsoon and monsoon seasons.

21416

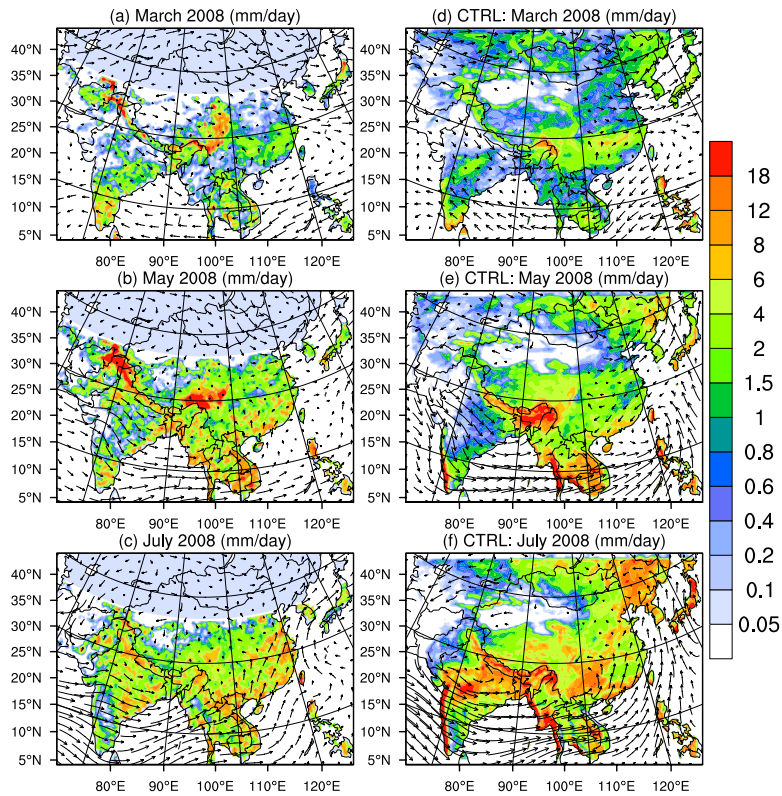


Fig. 5. Wind fields at 850 hPa and precipitation for March (a and d), May (b and e), and July (c and f) 2008. (a), (b), and (c) are the results from the ERA-Interim reanalysis and the TRMM rainfall measurement. (d), (e), and (f) are the results from the CTRL experiment.

21417

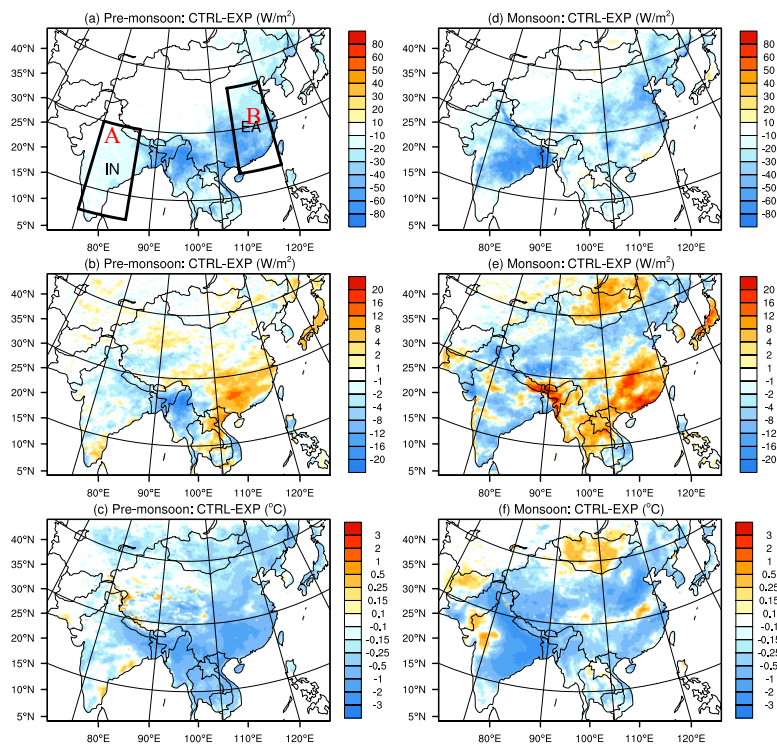


Fig. 6. Simulated changes in downward shortwave radiation reaching the surface (a and d), outgoing longwave radiation at the top (b and e), and surface temperature (c and f) in the pre-monsoon (a, b and c) and monsoon seasons (d, e and f). India (IN) and Eastasia (EA) are highlighted in (a). A and B in (a) are the two sites used in Fig. 12.

21418

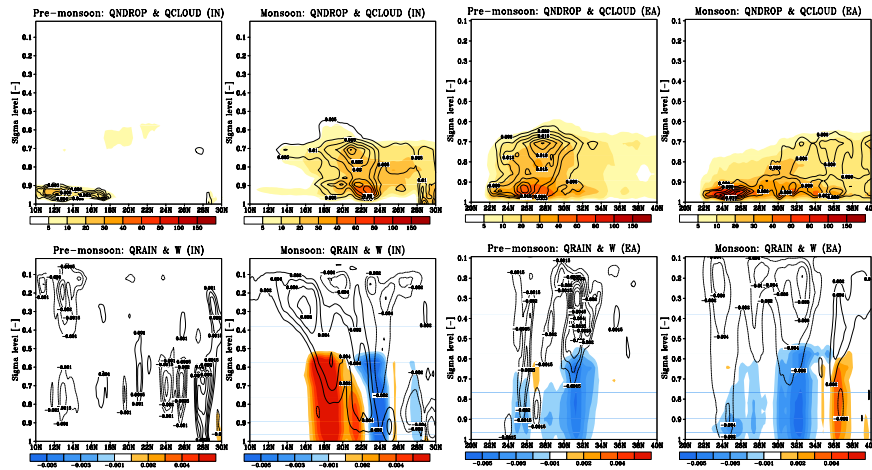


Fig. 7. Vertical cross-sections of cloud-related variables over IN and EA (shown in Fig. 6a). Color and lines on the top panel represent cloud water mixing ratio (QCLOUD) and cloud droplet number concentrations (QNDROP). Color and lines on the bottom panel represent rain water mixing ratio (QRAIN) and vertical velocity (W).

21419

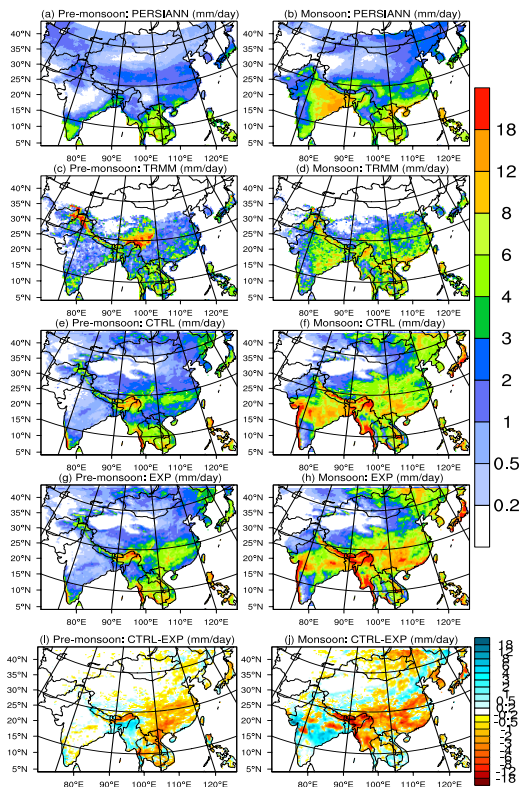


Fig. 8. Pre-monsoon and monsoon precipitation from satellite (PERSIANN and TRMM) and model experiments (CTRL and EXP). The bottom panel shows the difference in pre-monsoon and monsoon precipitation between CTRL and EXP.

21420

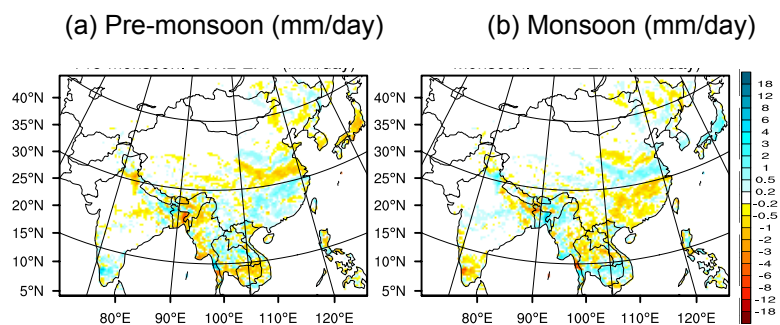


Fig. 9. Differences in pre-monsoon (a) and monsoon (b) precipitation between the experiments with dust aerosols and without dust aerosols.

21421

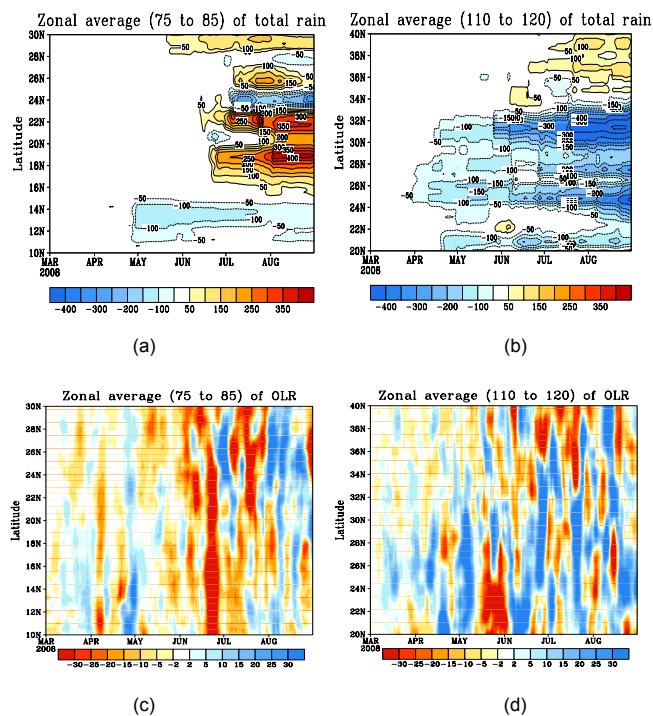


Fig. 10. Evolution of precipitation (latitude-time cross-sections of mean precipitation along 75–85° E (a) and 110–120° E (b); (c) and (d) are outgoing longwave radiation along 70–80° E and 110–120° E.

21422

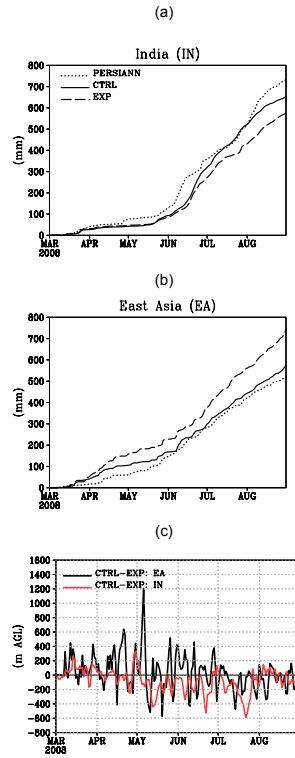


Fig. 11. Accumulative precipitation over the simulation period from PERSIANN, CTRL, and EXP over IN **(a)** and EA **(b)**. **(c)** shows differences in lifting condensation level (LCL) between CTRL and EXP over the two regions.

21423

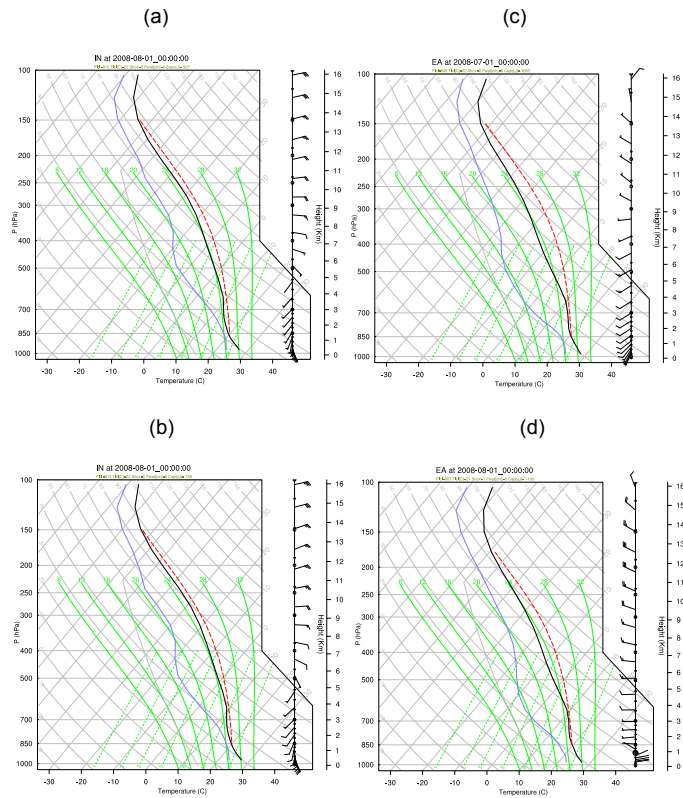


Fig. 12. Sounding plots in August 2008 at Site A (23° N, 95° E) **(a and b)** in IN and Site B (30° N, 120° E) **(c and d)** in EA (shown in Fig. 6a). Blue and black lines represent T_d (dew point temperature) and T (temperature). Red dashed shows the air parcel path.

21424

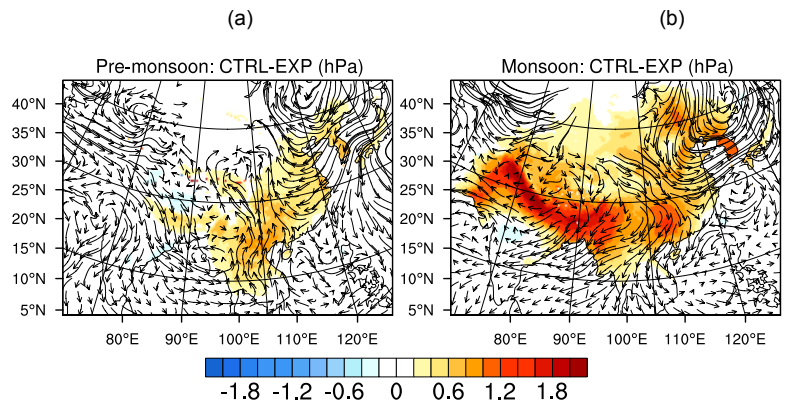


Fig. 13. Simulated changes in sea level pressure (SLP) and wind vectors at 850 hPa in the pre-monsoon **(a)** and monsoon **(b)** seasons.