Impacts of Asian summer monsoon on seasonal and interannual variations of aerosols over eastern China

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[1] China is located in a large monsoon domain; variations in meteorological fields associated with the Asian summer monsoon can influence transport, deposition, and chemical reactions of aerosols over eastern China. We apply a global three-dimensional Goddard Earth Observing System (GEOS) chemical transport model (GEOS-Chem) driven by NASA/GEOS-4 assimilated meteorological data to quantify the impacts of the East Asian summer monsoon on seasonal and interannual variations of aerosols over eastern China. During the summer monsoon season, four channels of strong cross-equatorial flows located within 40°E–135°E are found to bring clean air to China from the Southern Hemisphere. These channels have the effect of diluting aerosol concentrations in eastern China. In the meantime, rain belts associated with the summer monsoon move from southeastern to northern China during June–August, leading to a large wet deposition of aerosols. As a result, aerosol concentrations over eastern China are the lowest in summer. Sensitivity studies with no seasonal variations in emissions indicate that the Asian summer monsoon can reduce surface layer $PM_{2.5}$ (particles with a diameter of 2.5 μm or less) aerosol concentration averaged over eastern China (110°E–120°E, 20°N–45°N) by about 50–70%, as the concentration in July is compared to that in January. We also compare simulated $PM_{2.5}$ concentrations in the weak monsoon year of 1998 with those in the strong monsoon year of 2002, assuming same emissions in simulations for these 2 years. Accounting for sulfate, nitrate, ammonium, black carbon, organic carbon, as well as submicron mineral dust and sea salt, surface layer $PM_{2.5}$ concentration averaged over June–August and over eastern China is 7.06 $\mu g\ m^{-3}$ (or 44.3%) higher in the weak monsoon year 1998 than in the strong monsoon year 2002, and the column burden of $PM_{2.5}$ is 25.1 mg m^{-2} (or 73.1%) higher in 1998 than in 2002. As a result, over eastern China, the difference in summer aerosol optical depth between 1998 and 2002 is estimated to be about 0.7. These results have important implications for understanding air quality and climatic effects of aerosols in eastern China.

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1. Introduction

[2] Aerosols are major air pollutants in the atmosphere. They also influence climate change directly by absorbing and scattering radiation and indirectly by changing cloud properties [*Intergovernmental Panel on Climate Change* (*IPCC*), 2007]. With the rapid economic development in China over the past two decades, concentrations of aerosols and aerosol precursors are now among the highest in the world [*van Donkelaar et al.*, 2006; *X.-Y. Zhang et al.*, 2007]. Understanding the roles of aerosols over China in air quality and climate change requires a full understanding of their seasonal and interannual variations.

[3] China is located within the largest monsoon (Asian monsoon) domain in the world. In May–September every year, the Asian summer monsoon prevails; strong cross-equatorial winds flow from the Southern Hemisphere (SH) to the Northern Hemisphere (NH), bringing warm and moist air from the oceans to eastern China. Rain belts and associated deep convection that stretch for thousands of kilometers move from southeastern to northern China during this time period. In the months November to March, the Asian winter monsoon prevails; strong northerlies in high latitudes bring cold and dry air to eastern China. While such seasonal variations are about the same every year, the strength of the Asian monsoon exhibits large interannual

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variations as a result of the interactions between atmosphere and oceans [*Webster et al.*, 1998]. These seasonal and interannual variations in meteorological fields associated with the Asian monsoon can influence aerosols over eastern China by influencing transport, chemical reactions, and deposition of aerosols.

[4] Measurements have suggested that Asian summer monsoon is influential on seasonal variations of aerosols. Ground measurements in a number of cities over eastern China showed that aerosol concentrations are generally the lowest in summer [*Qin et al.*, 1998; *He et al.*, 2001; *Ye et al.*, 2003; *Dan et al.*, 2004; *Sun et al.*, 2004; *Wang et al.*, 2005; *Duan et al.*, 2006; *Cao et al.*, 2007]. Measurements of meteorological parameters and aerosols at the Maldives in the Indian Ocean indicated that clean air from the SH leads to low aerosol concentrations at the Maldives in the wet summer monsoon season [*Corrigan et al.*, 2006]. Although these studies suggested that the Asian summer monsoon has a large impact on aerosols, no previous studies, to our knowledge, have quantified the impact of the Asian summer monsoon on seasonal and interannual variations of aerosols over eastern China.

[5] Compared to the studies that have examined the impacts of the Asian summer monsoon on aerosols, more attention has been paid to study the role of aerosols in influencing the Asian summer monsoon. *Menon et al.* [2002] reported that atmospheric heating by increasing concentrations of black carbon aerosol in Asia might be responsible for the "southern flooding and northern drought" in China, the pattern of changes in rainfall associated with the weakening of the Asian summer monsoon since the late 1970s. *Ramanathan et al.* [2005] showed using a general circulation model that global dimming by the scattering and absorption of aerosols led to a long-term (multidecadal) weakening of the South Asian monsoon by altering the meridional temperature gradient between the Asian land mass and the Indian Ocean. Recently, *Lau et al.* [2006] reported that the absorbing aerosols (mineral dust and black carbon) can intensify the Indian monsoon through the so-called "Elevated-Heat-Pump" effect. They found based on observation that anomalously high concentrations of absorbing aerosols during the premonsoon season can lead to the formation of a warm-core upper level anticyclone over the Tibetan Plateau, influencing the development of the monsoon and the subsequent monsoon rainfall in India [*Lau and Kim*, 2006].

[6] Because of the two-way aerosol-monsoon interactions mentioned above, quantifying the spatial distribution and temporal variation of aerosols in China is very important for studies of air quality and climate change. In this work we assess the impacts of Asian summer monsoon on seasonal and interannual variations of aerosols in eastern China using the global chemical transport model GEOS-Chem driven by the assimilated meteorological fields. We give a brief description of the GEOS-Chem model and all the numeral experiments in section 2. Section 3 presents the simulated seasonal variation of aerosols in eastern China. Section 4 identifies the major factors associated with the Asian summer monsoon that have large impacts on seasonal variation of aerosols in China. In section 5 we quantify the impact of the Asian summer monsoon on interannual variation of aerosols by comparing aerosol concentrations over eastern China simulated in a weak monsoon year with those simulated in a strong monsoon year. Sections 3–5 focus on anthropogenic aerosols. Section 6 examines seasonal and interannual variations of aerosols in eastern China with further consideration of mineral dust and sea salt aerosol.

2. Model Description and Numerical Experiments

2.1. GEOS-Chem Model

[7] We simulate aerosols using the global chemical transport model GEOS-Chem (version 7.3.6, http://acmg.seas.harvard.edu/geos) driven by the assimilated meteorological fields from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). All the simulations are driven by GEOS-4 meteorological fields with a horizontal resolution of 4° latitude by 5° longitude. The original vertical resolution of GEOS-4 meteorological fields is 55 levels, extending from the surface up to 0.01 hPa. In order to minimize the amount of memory required to run GEOS-Chem, we select to run with a reduced vertical resolution of 30 levels (the original layers above ~50 hPa are lumped together online). The GEOS-Chem model includes a fully coupled treatment of tropospheric ozone-NO_x-VOC chemistry and sulfate (SO_4^{2-}, nitrate NO_3^-, ammonium (NH_4^+), organic carbon (OC), black carbon (BC), mineral dust, and sea salt aerosols [*Park et al.*, 2003, 2004; *Alexander et al.*, 2005; *Fairlie et al.*, 2007]. Simulation of BC and OC in GEOS-Chem is described by *Park et al.* [2003]. A 10% carbon yield of OC from biogenic terpenes [*Park et al.*, 2003] is applied to global terpene emission inventory that depends on vegetation type, monthly adjusted leaf area index, and temperature [*Guenther et al.*, 1995]. The implementation of sulfate/nitrate/ammonium is described by *Park et al.* [2004]. Partitioning of total ammonia and nitric acid between the gas and aerosol phases is calculated using the ISORROPIA thermodynamic equilibrium model [*Nenes et al.*, 1998]. Aerosol and gas phase simulations are coupled through formation of sulfate and nitrate, heterogeneous chemistry, and aerosol effects on photolysis rates [*Martin et al.*, 2003]. Heterogeneous reactions include hydrolysis of N_2O_5 on different types of aerosols [*Evans and Jacob*, 2005] and irreversible absorption of NO_3, NO_2 and HO_2 on wet aerosols [*Jacob*, 2000]. The model simulates submicron and supermicron sea salt [*Alexander et al.*, 2005], and dust aerosol in four size bins (0.1–1.0, 1.0–1.8, 1.8–3.0, and 3.0–6.0 μm dry radius) [*Fairlie et al.*, 2007].

2.2. Dry and Wet Deposition

[8] The aerosol wet deposition scheme in the GEOS-Chem follows that of *Liu et al.* [2001], which includes scavenging in convective updrafts, rainout and washout from convective anvils and large-scale precipitation, and return of chemical species to the atmosphere following evaporation. It has been extended to predict wet deposition of soluble gases on the basis of effective Henry's law partitioning in warm clouds, retention efficiency upon droplet freezing in mixed clouds, and surface coating or condensation of ice crystals in cold clouds [*Mari et al.*, 2000]. For the scavenging of aerosols, sulfate, ammonium, nitrate, and

Table 1. Summary of Annual Emissions of Aerosols and Aerosol Precursors in Asia and Eastern China[a]

Species	Asia	Eastern China
SO_2 (Tg S yr^{-1})	26.55	11.79
aircraft	0.01	<0.01
anthropogenic	23.40	11.68
biomass burning	0.33	<0.01
biofuel	0.16	0.05
volcanoes	1.96	0.00
ship	0.69	0.05
NH_3 (Tg N yr^{-1})	21.58	5.24
anthropogenic	16.40	4.72
natural	2.87	0.30
biomass burning	1.50	0.03
biofuel	0.81	0.20
NO_x (Tg N yr^{-1})	16.87	5.77
aircraft	0.09	0.01
anthropogenic	10.95	5.01
biomass burning	1.78	0.03
biofuel	1.31	0.35
fertilizer	0.21	0.05
lightning	1.34	0.11
soil	1.18	0.20
OC (Tg C yr^{-1})	16.65	2.86
anthropogenic	4.63	1.58
biomass burning	6.18	0.11
biofuel	3.75	0.99
biogenic	2.09	0.17
BC (Tg C yr^{-1})	4.60	1.58
anthropogenic	2.84	1.29
biomass burning	0.75	0.01
biofuel	1.01	0.28

[a]Asia domain: 60°E–155°E, 10°N–55°N; eastern China: 110°E–120°E, 20°N–45°N.

hydrophilic OC and BC aerosols are assumed to be fully soluble. Hydrophobic OC and BC are assumed to be insoluble. Following *Cooke et al.* [1999], we assume that ambient conversion of OC and BC from hydrophobic to hydrophilic occurs with an exponential decay lifetime of 1.15 days. Dry deposition of aerosols and gases is based on a standard resistance-in-series model dependent on local surface type and meteorological conditions [*Wesely*, 1989] and implemented as described by *Wang et al.* [1998].

2.3. Emissions

[9] Global emissions of ozone precursors, aerosol precursors, and aerosols in the GEOS-Chem generally follow *Park et al.* [2003, 2004], except that anthropogenic emissions (including emissions from power, industry, residential, and transportation) in the Asian domain are updated to David Streets' 2006 emission inventory (http://www.cgrer.uiowa.edu/EMISSION_DATA_new/index_16.html). The monthly variation of emissions from each emission sector in China is taken from *Q. Zhang et al.* [2007]. Table 1 summarizes the annual emissions of SO_2, NH_3, NO_x, BC, and OC in the entire Asian domain (60°E–155°E, 10°N–55°N) and eastern China (110°E–120°E, 20°N–45°N). Monthly variations of emissions (anthropogenic plus natural emissions) of these species averaged over eastern China are shown in Figure 1. Emissions of SO_2, BC, and OC are the highest in winter months because of winter energy use. Emissions of NO_x are slightly higher in summer than in winter, because of the relatively larger emissions from lightning and soil in summer. Emissions of NH_3 are the highest in summer, because NH_3 from sources such as fertilizer application and domesticated animals are the maximum at high temperatures [*Aneja et al.*, 2000; *Park et al.*, 2004]. Emission schemes for sea salt and mineral dust follow those of *Alexander et al.* [2005] and *Fairlie et al.* [2007], respectively, with emissions calculated based on the GEOS meteorological fields.

2.4. Numerical Experiments

[10] Concentrations of aerosols are determined by both emissions and meteorological fields. For the purpose of this work, we perform five numerical experiments using the GEOS-Chem model: (1) 2001EmAnn is a control simulation driven by the GEOS-4 meteorological fields for the period of 1 January to 31 December of year 2001, allowing monthly variation of emissions of all species; (2) 2001EmJan is a simulation that is the same as 2001EmAnn, except that anthropogenic emissions of all species are fixed at January values throughout the simulation for year 2001; (3) 2001EmJul is a simulation that is the same as 2001EmAnn, except that anthropogenic emissions of all species are fixed at July values throughout the simulation for year 2001; (4) 1998EmAnn is a simulation that is the same as 2001EmAnn, except that the simulation is driven by the GEOS-4 meteorological fields for the period of 1 January to 31 December of year 1998; and (5) 2002EmAnn is a simulation that is the same as 2001EmAnn, except that the simulation is driven by the GEOS-4 meteorological fields for the period of 1 January to December 31 of year 2002.

[11] Simulations 1–3 are performed for year 2001, because 2001 is an ordinary year in terms of monsoon strength [*Li and Zeng*, 2002, 2003, 2005]. Since anthropogenic emissions in eastern China are the highest in winter and lowest in summer [*Q. Zhang et al.*, 2007], by fixing emissions at January values in 2001EmJan or at July values in 2001EmJul, seasonal variations of aerosols simulated in 2001EmJan and 2001EmJul represent the influences of changing meteorological fields associated with the Asian summer monsoon. Note that emissions outside Asia domain

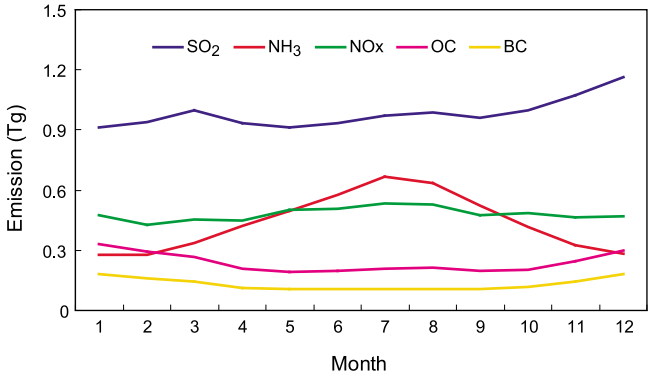

Figure 1. Monthly variations in emissions of SO_2 (Tg S $month^{-1}$), NH_3 (Tg N $month^{-1}$), NO_x (Tg N $month^{-1}$), OC (Tg C $month^{-1}$), and BC (Tg C $month^{-1}$) over eastern China. Values shown are the sum of anthropogenic and natural emissions, with sources of emissions listed in Table 1. Natural emissions are estimated based on the GEOS-4 meteorological fields of 2001.

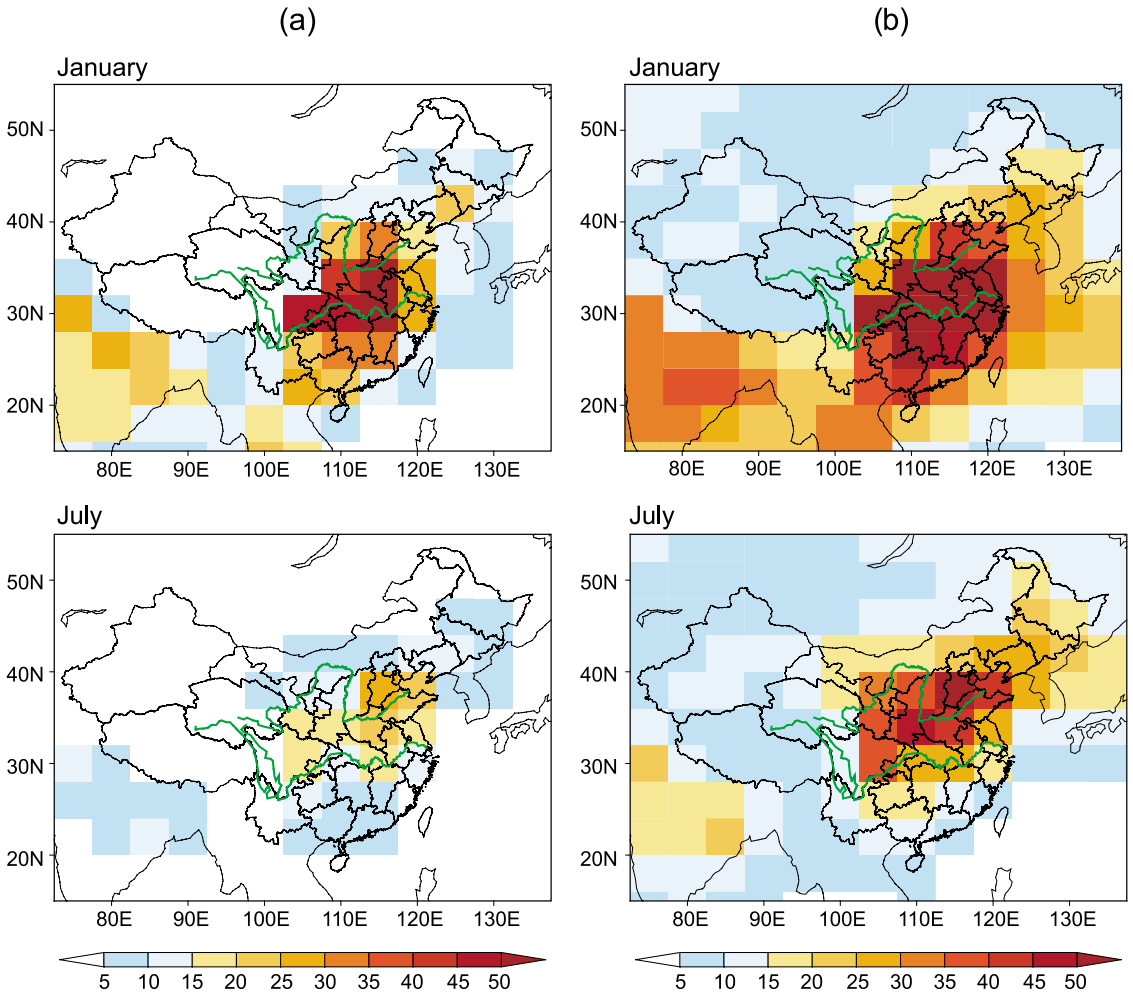

Figure 2. Simulated (a) surface layer (~1000 hPa) concentrations (μg m^{-3}) and (b) column burdens (mg m^{-2}) of $PM_{2.5}$ for (top) January and (bottom) July of 2001. $PM_{2.5}$ is the sum of sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols.

have monthly variations and are kept the same in all the simulations in this work.

[12] Meteorological fields are quite different in magnitude between strong and weak monsoon years. The Asian summer monsoon can be divided into two independent subsystems, the East Asian summer monsoon (EASM) and the South Asian summer monsoon (SASM) [*Zhu et al.* 1986; *Tao and Chen*, 1987; *Chen et al.*, 1991; *Li and Zeng*, 2002; *Chen et al.*, 2004; *Qian et al.*, 2004]. A number of monsoon indices have been designed to quantify the year-to-year variation in the intensity of EASM and SASM [*Wang and Fan*, 1999; *Li and Zeng*, 2002, 2003; *Wang et al.*, 2008]. Abundant rainfall within the local rain-bearing monsoon system is considered to be a strong monsoon for SASM. However, for the summer monsoon in China (EASM), a strong summer monsoon is characterized by strong southerlies extending from southern to northern China, a deficit of rainfall in the middle and lower reach of the Yangtze River, and large rainfall in northern China, with the pattern of rainfall associated with the path of southerlies that bring moist air from the oceans [*Li and Zeng*, 2002; *Wang et al.*, 2008]. On the contrary, a weak summer monsoon in China has weak southerlies that cannot push the monsoon system northward enough, resulting in a large rainfall in southern China and a deficit of rainfall in northern China. According to the monsoon index defined by *Li and Zeng* [2002, 2003, 2005], year 1998 is a weak monsoon year and year 2002 is a strong monsoon year for both EASM and SASM. Simulations 1998EmAnn and 2002EmAnn are hence designed to assess the influence of summer monsoon strength on predicted aerosol concentrations in eastern China.

[13] Considering that sea salt is not a dominant aerosol species in China, and simulations of mineral dust aerosol generally have larger uncertainties than simulations of anthropogenic aerosols [*Liao et al.*, 2004; *Fairlie et al.*, 2007], we present our model results accounting for sulfate, nitrate, ammonium, BC, and OC in sections 3–5, and present model results with further consideration of sea salt and dust in section 6.

3. Predicted Distribution and Seasonal Variation of Aerosols Over China

[14] Figure 2a shows the simulated surface layer (~1000 hPa) $PM_{2.5}$ (the sum of sulfate, nitrate, ammonium, BC, and OC

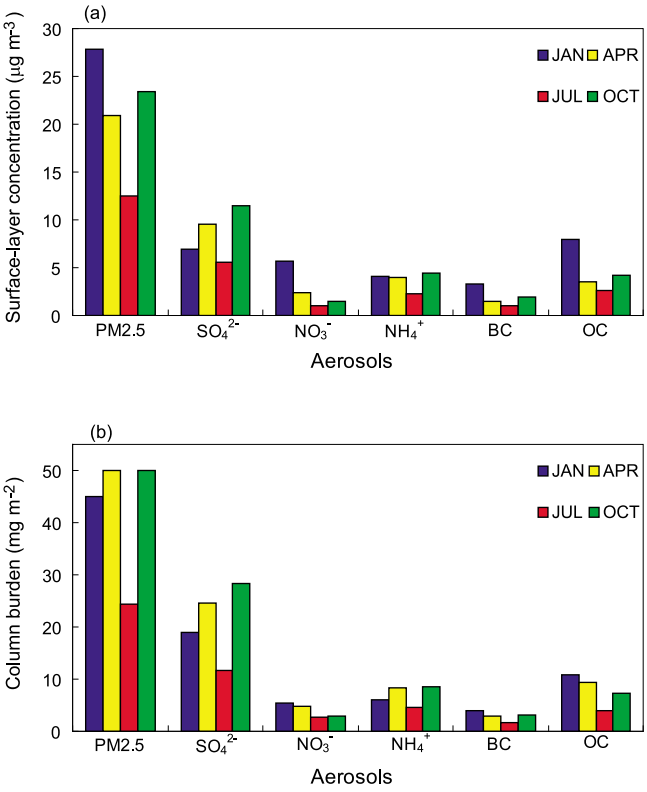

Figure 3. Simulated (a) surface layer concentrations (μg m^{-3}) and (b) column burdens (mg m^{-2}) of $PM_{2.5}$, NO_3^-, SO_4^{2-}, NH_4^-, BC, and OC averaged over eastern China (110°E–120°E, 31°N–45°N) for January, April, July, and October of year 2001. $PM_{2.5}$ accounts for sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols.

aerosols) concentrations from simulation 2001EmAnn for months January and July of year 2001. $PM_{2.5}$ concentrations are high over eastern China (110°E–120°E, 20°N–45°N) throughout the year, with concentrations exceeding 50 μg m^{-3} in January and 15–25 μg m^{-3} in July.

[15] Figure 3a shows simulated concentrations of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC that are averaged over eastern China (110°E–120°E, 31°N–45°N) for January, April, July, and October of year 2001. Concentrations of $PM_{2.5}$, NO_3^-, BC, and OC are the highest in January and the lowest in July. Sulfate exhibits a different seasonal variation of having the maximum concentration in October, which can be explained by the net effect of photochemical reactions and wet deposition. While stronger photochemical reactions can lead to more sulfate formation in July than in October, more precipitation leads to larger wet deposition of sulfate in July in eastern China. Ammonium aerosol exists as ammonium bisulfate, ammonium sulfate, or ammonium nitrate. While ammonium concentration is the lowest in July because of the summer monsoon rainfall, its concentrations are about the same (~4.5 μg m^{-3}) in January, April, and October as a result of the different seasonal variations of sulfate and nitrate. In the surface layer, sulfate is predicted to be the most abundant aerosol species over eastern China, followed by OC, NH_4^+, NO_3^- and BC. Simulated surface layer $PM_{2.5}$ concentration in January is about three times the value in July over eastern China.

[16] Simulated $PM_{2.5}$ concentrations are compared with measurements at different sites in China in Table 2. Because of the lack of long-term measurements in China, seasonal mean concentrations observed in different years are listed in Table 2 to show the general magnitude and seasonal features of measured concentrations. The model generally underestimates $PM_{2.5}$ concentrations at all sites, but captures well the seasonal trend of high concentrations in winter and low levels in summer. The underprediction of concentrations can be explained by our coarse horizontal resolution of 4° latitude by 5° longitude that has an effect of averaging the concentrations over the large grid cell, and by the fact that most measurements shown in Table 2 were taken in heavily polluted cities. We do not consider mineral dust and sea salt aerosols here, which also contribute to the low bias in model predictions. Model predictions with dust and sea salt will be presented in section 6.

[17] Figure 2b shows the simulated distributions of column burden of $PM_{2.5}$ for January and July of year 2001. Similar to surface layer concentrations, column burdens of $PM_{2.5}$ over eastern China are higher in January than in July. Because of the large contribution of sulfate to $PM_{2.5}$, the column burden of $PM_{2.5}$ in January is lower than the burdens in April and October (Figure 3b).

4. Impact of Asian Summer Monsoon on Seasonal Variation of Aerosols

[18] The Asian summer monsoon is characterized by strong cross-equatorial flows and convection, and precipitation. We examine in this section the impacts of major factors associated with the Asian summer monsoon on aerosol concentrations over eastern China.

4.1. Cross-Equatorial Flows

[19] Cross-equatorial flows from the SH to NH in the Asian summer are a dominant feature of the lower tropospheric monsoon circulation [*Bunker*, 1965; *Joseph and Raman*, 1966; *Findlater*, 1969]. The strength and location of the cross-equatorial flows have interannual variations [*Chen et al.*, 1991]. Figure 4a shows the 925 hPa stream lines and wind velocity averaged over June-July-August (JJA) of year 2001. Strong cross-equatorial flows exist in the tropical monsoon regions and can eventually arrive in eastern China. For example, the strongest flows located between 40°E and 65°E at the equator can pass the Arabian Sea, Indian Peninsula, the Bay of Bengal, Indochina peninsula, the South China Sea, and then reach southern China.

[20] The strength and location of cross-equatorial flows can be identified by the time-longitude distribution of 925 hPa meridional wind averaged over 5°S–5°N, as shown in Figure 4b. Four channels of cross-equatorial flows exist in the Asian monsoon domain, covering the longitude ranges of 40°E–65°E, 85°E–95°E, 105°E–115°E, and 120°E–135°E. The first channel in 40°E–65°E has strong winds in April–October, while the other three channels are strong in May–August. *Corrigan et al.* [2006] have shown that the flows from the SH during summer monsoon season reduces aerosol concentrations in the Maldives (6.8°N, 73.2°E).

Table 2. Comparisons of Observed and GEOS-Chem $PM_{2.5}$ Concentrations

	Concentrations ($\mu g\ m^{-3}$)				
	Summer		Winter		
Location	GEOS-Chem	Observed	GEOS-Chem	Observed	References
Beijing (116.5°E, 39.9°N)	24.4	117.2 ± 48.3	57.5	126.5 ± 66.1	*Cao et al.* [2007]
				175.9	*He et al.* [2001]
		104.1 ± 45.1		257.6 ± 85.8	*Dan et al.* [2004]
		77.3 ± 55.7		135.7 ± 96.6	*Sun et al.* [2004]
		93.29 ± 56.26		214.23 ± 159.34	*Wang et al.* [2005]
		88.99		122.09	*Duan et al.* [2006]
		107 ± 61		182 ± 107	*Zhang et al.* [2006]
		71		108	*Xu et al.* [2007]
		59.2			*Wu and Wang* [2007]
Changchun (125.2°E, 45.5°N)	9.7	59.6 ± 21.9	45.6	140.5 ± 28.6	*Cao et al.* [2007]
Qingdao (120.2°E,36.0°N)	21.3	30.1 ± 16.1	31.8	127.9 ± 58.5	*Cao et al.* [2007]
Tianjin (117.1°E, 39.0°N)	24.4	103.2 ± 27.9	57.5	179.4 ± 87.8	*Cao et al.* [2007]
Xi'an (108.6°E, 34.2°N)	17.3	130.8 ± 58.5	47.6	375.2 ± 143.5	*Cao et al.* [2007]
Yulin (109.5°E, 38.2°N)	10.9	50.5 ± 23.2	31.8	150.6 ± 77.3	*Cao et al.* [2007]
Chongqing (106.3°E, 29.4°N)	17.5	116.3 ± 38.1	44.0	311.8 ± 114.1	*Cao et al.* [2007]
Guangzhou (113.1°E, 23.1°N)	4.2	78.1 ± 29.7	14	105.9 ± 71.4	*Cao et al.* [2003, 2004]
		49.1 ± 9.3		156.0 ± 93.6	*Cao et al.* [2007]
Hong Kong (115.1°E, 25.2°N)	3.4	31 ± 16.9	9.0	54.5 ± 22.9	*Cao et al.* [2003, 2004]
		40.1 ± 14.0		64.0 ± 29.6	*Cao et al.* [2007]
Hangzhou (120.1°E, 30.2°N)	14.5	90.6 ± 40.8	25.5	168.6 ± 54.6	*Cao et al.* [2007]
Shanghai (121.3°E, 31.1°N)	16.0	52.2 ± 19.4	28.5	151.1 ± 95.4	*Cao et al.* [2007]
		35.85		93.91	*Ye et al.* [2003]
		59.7			*Wu and Wang* [2007]
Nanjing (118.5°E, 32.0°N)	16.0	69.1	28.5	139.5	*Huang et al.* [2006]
Wuhan (114.2°E, 30.4°N)	15.6	70.8 ± 21.3	51.6	166.6 ± 72.7	*Cao et al.* [2007]
Xiamen (118.0°E, 24.3°N)	4.7	25.2 ± 15.8	17.5	70.2 ± 32.2	*Cao et al.* [2007]
Shenzhen (114.1°E, 24.3°N)	3.4	47.1 ± 16.7	9.0	60.8 ± 18.0	*Cao et al.* [2003, 2004]
Zhuhai (113.3°E, 22.2°N)	3.4	31.0 ± 20.0	9.0	59.3 ± 23.7	*Cao et al.* [2003, 2004]

[21] To quantify the impact of each channel of cross-equatorial flows on aerosols over eastern China, a cross-equatorial flow index I_c is defined for each channel following the study of *Wang and Fu* [2002],

$$I_c = \sum_{ij} v_{ij} s_{ij} \Big/ \sum_{ij} s_{ij}, \quad (1)$$

where v_{ij} and s_{ij} are the 950 hPa meridional wind and the area of grid cell (i,j), respectively. Here i represents the gird cells located in the longitude range of each of the four channels mentioned above and j represents the grid cells over 5°S–5°N. The correlation of daily I_c of each channel with simulated daily $PM_{2.5}$ concentration at each grid cell is then calculated over April–July for the first channel in 40°E–65°E and over May–July for the other three (Figure 5), with the time periods for correlation calculation determined by the months with increasing strength of meridional winds. As shown in Figure 5a, correlations between $PM_{2.5}$ concentrations and I_c of the first channel located over 40°E–65°E are generally negative, both along the pathway and over eastern China, indicating that this channel of flows has an effect of reducing $PM_{2.5}$ concentrations in eastern China. An area of positive correlation is found in southwestern China under the influence of the second channel of flows in 85°E–95°E (Figure 5b); although this channel of cross-equatorial flows reduces aerosol concentrations in a large fraction of the tropical monsoon regions, it increases $PM_{2.5}$ concentrations in southwestern China by bringing pollutants from South Asia. Meanwhile, this channel also reduces $PM_{2.5}$ concentrations in the Yangtze River basin (30°N) in eastern China. The third and fourth channels of cross-equatorial flows mainly reduce aerosol concentrations over eastern China (Figures 5c and 5d, respectively). These results indicate that cross-equatorial flows associated with the Asian summer monsoon have different impacts on the spatial distributions of aerosols in China, and the overall effect of these flows is to reduce aerosol concentrations in summer over eastern China.

4.2. Summer Monsoon Rainfall

[22] During the Asian summer monsoon, under the influence of the western Pacific subtropical high, rain belts that stretch thousands of kilometers in the west-east direction in eastern China undergo three standing stages and two abrupt northward shifts every year [*Tao and Chen*, 1987; *Lau et al.*, 1988; *Ding*, 1992; *Tanaka*, 1992, *Wang and Ding*, 2008]. The rain belts of the summer monsoon firstly appear over southern China from April to mid-June, intensify and shift to the middle and lower reach of the Yangtze River around mid-June and stay there until mid-July (the raining period of Meiyu). The second northward shift occurs in mid-July and the resulting rainfall in northern China lasts until mid-August.

[23] Negative correlation is found between precipitation and aerosol concentrations when we examine the predicted changes in pentad mean tropospheric column burdens of $PM_{2.5}$ from P31 (31 May to 4 June) to P36 (25–29 June) (Figure 6a). High column burdens of exceeding 50 mg m^{-2} are found over eastern China in the first three pentads (P31–

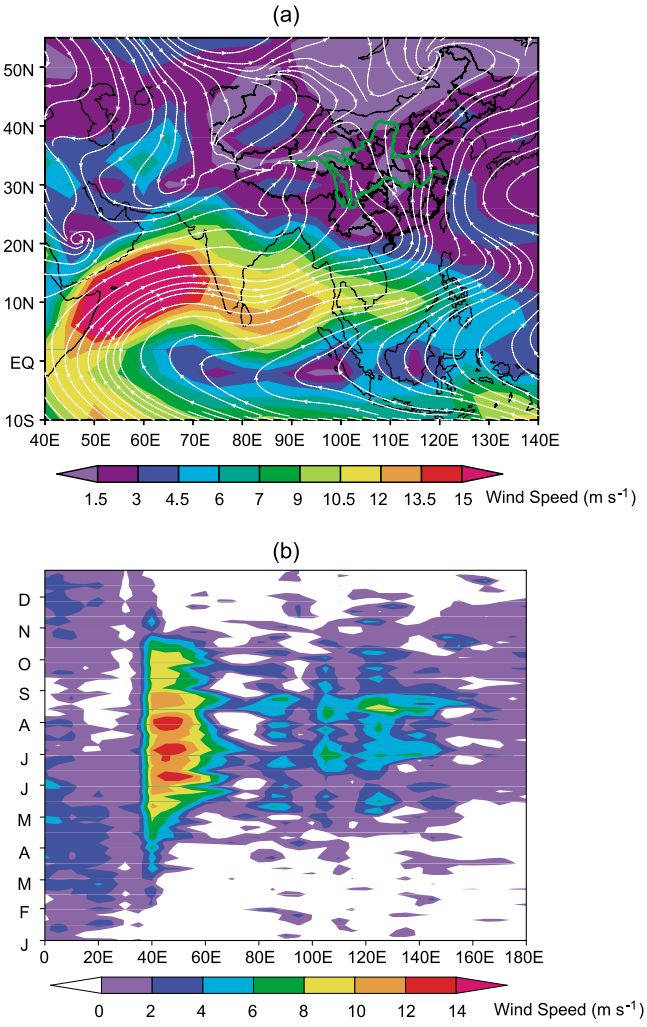

Figure 4. (a) The 925 hPa stream lines and wind velocity (m s^{-1}) averaged over June-July-August (JJA) of year 2001. (b) Time-longitude distribution of 925 hPa meridional wind (m s^{-1}) averaged over 5°S–5°N for year 2001. Meteorological fields are from the GEOS-4 assimilated meteorological data.

P33). Aerosol concentrations show reductions of about 50% starting from P34 (15–19 June), because of the onset of large monsoon rainfall (Meiyu) in China in P34. As shown in Figure 6b, rain belts are mainly located over southern China in early June. Corresponding to the northeastward retreat of the western Pacific subtropical high, rain belts with large rainfall move from southeastern China to northern and northeastern China as the summer monsoon develops and moves northward in P34. Heavy rainfall (Meiyu) starts in eastern China in P34, and lasts until mid-July. The distributions of precipitation are consistent with predicted reductions in $PM_{2.5}$ concentration shown in Figure 6a. The large impact of monsoon rainfall can also be seen clearly in simulated wet deposition of aerosols (Figure 6c). The maximum wet deposition locates in southern China in P31. As rain belts move northward, another area with large wet deposition appears in northern and northeastern China in P34. Results indicate that aerosol concentrations in eastern China are sensitive to summer monsoon rainfall, movement of rain belts, and the amount of rainfall.

4.3. Effect of Monsoon Versus That of Emissions on Seasonal Variation of $PM_{2.5}$

[24] Seasonal variations of aerosols are determined by both seasonal variations of emissions and meteorological parameters. To separate the role of changing emissions, we use fixed January and July emissions in simulations 2001EmJan and 2001EmJul, respectively. Figure 7 presents simulated seasonal variations of all aerosol species in eastern China from experiments 2001EmAnn, 2001EmJan, and 2001EmJul. Compared to the results of 2001EmAnn, predicted concentrations of BC and OC are higher in 2001EmJan and lower in 2001EmJul throughout the year, as a result of the highest anthropogenic emissions of BC and OC in January and the lowest emissions of these species in July (Figure 1). In DJF, concentrations of nitrate and ammonium obtained in 2001EmJul are much higher than those from 2001EmAnn and 2001EmJan, because of the highest emission of NH_3 in July (Figure 1) and the low temperatures in DJF that favor ammonium nitrate formation. In all simulations, $PM_{2.5}$ concentrations over eastern China are the lowest in summer and the highest in winter, despite the differences in emissions. Results indicate that seasonal variations in emissions are not the major factors that influence seasonal variation of $PM_{2.5}$ aerosols in eastern China.

[25] We can further quantify the impact of the monsoon on seasonal variations of $PM_{2.5}$ by the reduction rate of $R = ([PM_{2.5}]_{Jul} - [PM_{2.5}]_{Jan})/[PM_{2.5}]_{Jan}$, where $[PM_{2.5}]_{Jan}$ and $[PM_{2.5}]_{Jul}$ represent monthly mean concentrations of $PM_{2.5}$ in January and July, respectively. Figure 8 shows the reduction rates from each of the simulations 2001EmAnn, 2001EmJan, and 2001EmJul. In 2001EmAnn, large reduction rates of exceeding 60% are found over eastern China, the western North Pacific, and South Asia. Although the seasonal variations in emissions are turned off in both 2001EmJan and 2001EmJul, the pattern of reduction rates obtained in each of these two sensitivity simulations is quite similar to that obtained in 2001EmAnn, indicating the major role of the Asian summer monsoon in influencing $PM_{2.5}$ aerosol seasonal variation in eastern China. Averaged over the whole domain of eastern China, the reduction rates by monsoon are −55.1%, −53.2%, and −72.8% in simulations of 2001EmAnn, 2001EmJan, and 2001EmJul, respectively.

[26] It should be noted that the effect of the monsoon quantified here includes the effects of some natural variations in meteorological fields such as higher temperature and stronger convections in summer than in winter. It is difficult to separate the effect of such natural variation from the impact of the monsoon on aerosols. However, since sulfate is the most abundant aerosol species in the atmosphere and the strong photochemical reactions in summer increase sulfate formation, $PM_{2.5}$ concentrations can be the highest in summer with high temperature and high boundary layer height in the absence of a strong impact of the monsoon. The seasonal variation of $PM_{2.5}$ aerosols in the eastern United States (i.e., high concentrations in summer and low concentrations in winter [*DeBell et al.*, 2006]) is an example of such a case. This suggests that the natural variations in meteorological parameters can increase aerosol concentra-

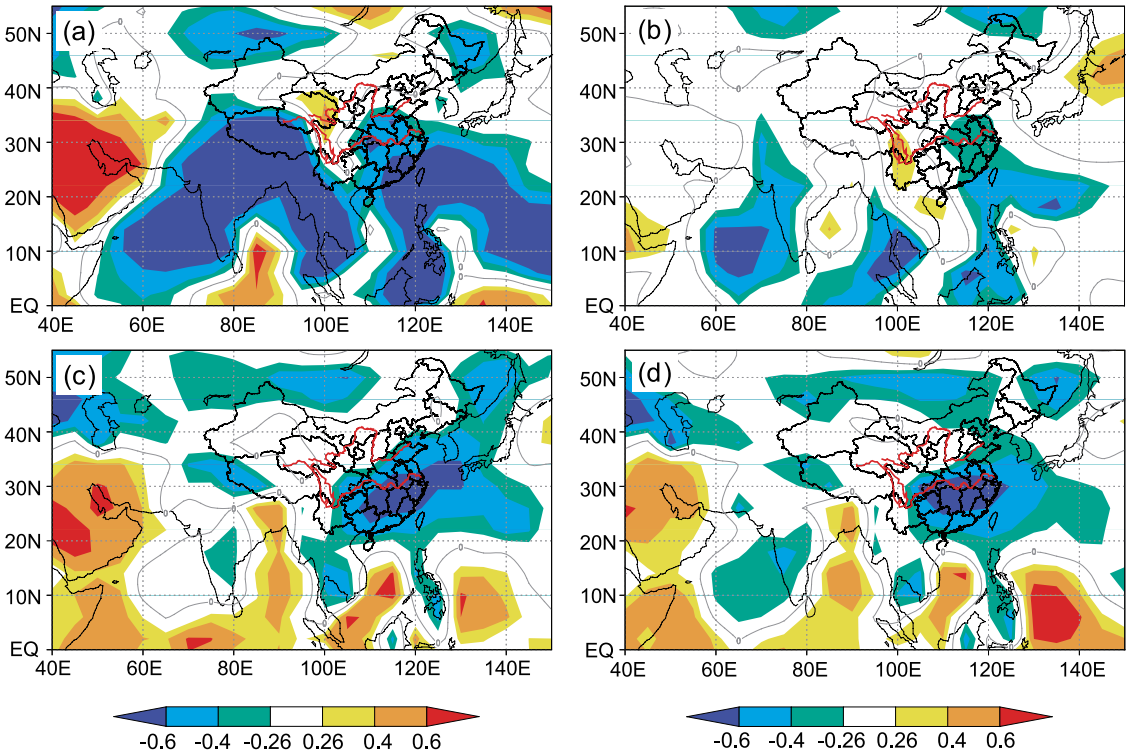

Figure 5. Correlation between simulated daily $PM_{2.5}$ concentrations and the cross-equatorial flow index I_c (equation (1)) for channels located at (a) 40°E–65°E, (b) 85°E–95°E, (c)105°E–115°E, and (d) 120°E–135°E. The correlation is calculated for the period of April–July for the channel located over 40°E–65°E and for the period of May–July for the other three channels. The colors denote the correlations significant at the 99% confidence level. See the text for the definition of I_c. $PM_{2.5}$ accounts for sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols.

tions in summer, which is opposite to the impact of Asian summer monsoon.

5. Impact of Asian Summer Monsoon Strength on Interannual Variations of Aerosols

[27] A strong summer monsoon in China (EASM) is characterized by strong southerlies extending from southern China to northern China, a deficit of rainfall in the middle and lower reaches of the Yangtze River, large rainfall in northern China, and movement of the rain belts associated with the strength of the southerlies. On the contrary, in a weak summer monsoon year, China experiences weak southerlies, large rainfall in southern China, and a deficit of rainfall in northern China. These features of either a weak or a strong monsoon year are summarized based on the anomalies in meteorological fields relative to climatological means. As mentioned in section 2.5, 1998 is a weak monsoon year and 2002 is a strong monsoon year according to the monsoon index defined by *Li and Zeng* [2002, 2003, 2005]. We examine in this section the differences in aerosol concentrations between 1998 and 2002.

5.1. Differences in Meteorological Fields Between Weak and Strong Monsoon Years

[28] Figure 9 shows the differences in precipitation, wind at 850 hPa, meridional wind at 925 hPa, and temperature at 1000 hPa between 1998 and 2002 for summer months June, July, and August. In all three months, South Asia had less precipitation of up to 4 mm d^{-1} in 1998 than in 2002, which has an effect of increasing local aerosol concentrations in 1998. Relative to 2002, heavier rainfall occurred over southern China and less precipitation was found over northern China in June of 1998, which agrees with the general features of weak and strong monsoon years. With the northward migration of monsoon rain belts, heavier rainfall occurred in 1998 over eastern China in July and over the Yangtze River basin (~30°N) in eastern China in August. On a seasonal mean basis, JJA precipitation averaged over eastern China was 1.16 mm d^{-1} higher in the weak monsoon year 1998 than in the strong monsoon year 2002.

[29] At 850 hPa altitude, stronger northwesterlies covered northern China in June of the weak monsoon year of 1998. This prevented the northward movement of monsoon rain belts, leading to larger rainfall in southern China. Stronger southwesterlies occurred at 850 hPa over eastern China in July and August, favoring the northward transport of pollutants. In the mean time, stronger northeasterlies occurred over the western North Pacific at 40°N in July, which prevented the outflow of pollutants from eastern China. Abnormal northerlies (stronger northerlies or weaker southerlies) occurred over northern China in August, which again was the typical behavior of winds in a weak monsoon year relative to a strong monsoon year. Similar behavior in

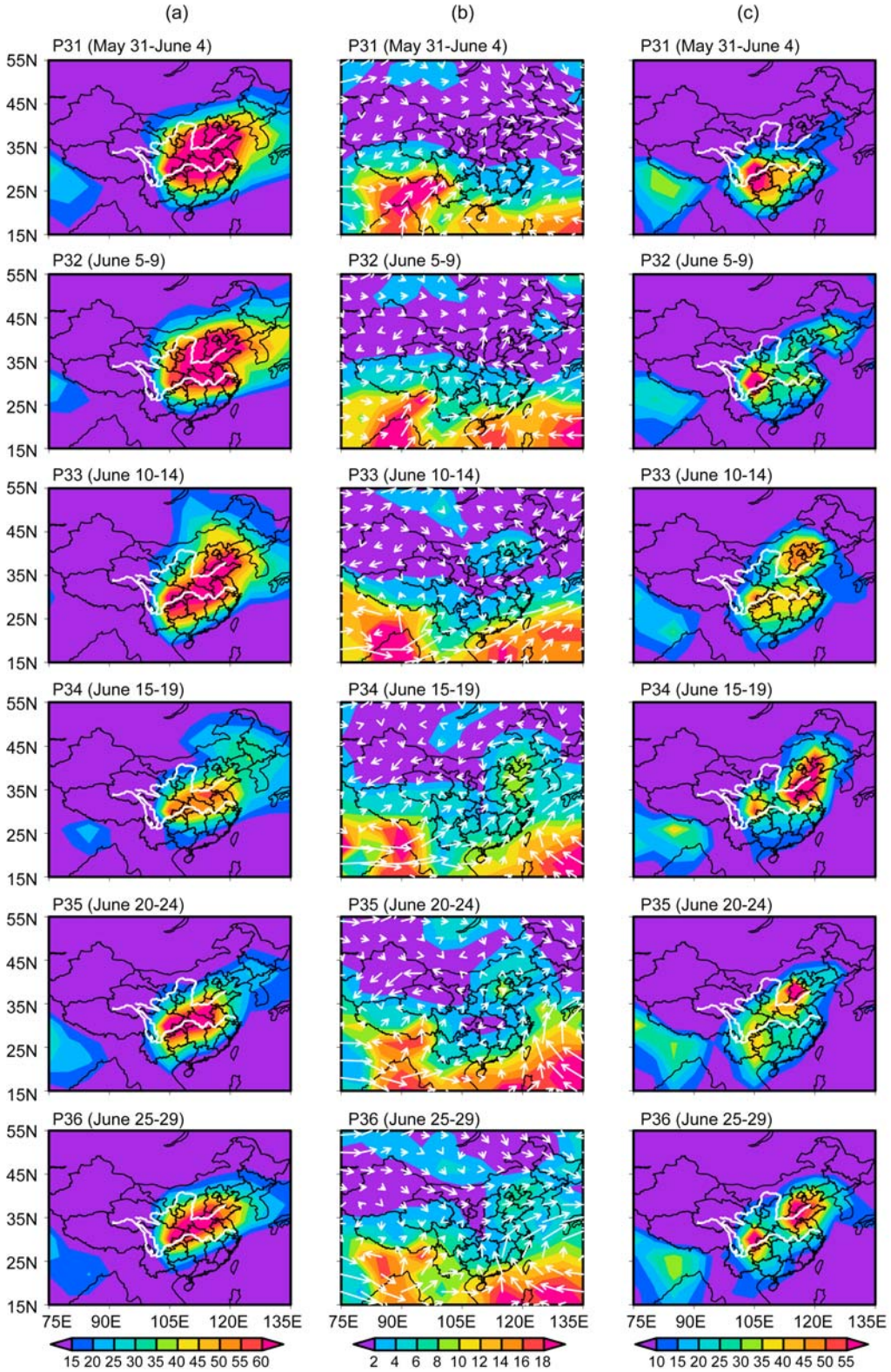

Figure 6. (a) Predicted pentad mean tropospheric column burdens (mg m^{-2}) of $PM_{2.5}$ from P31 (31 May to 4 June) to P36 (25–29 June). (b) The pentad mean rainfall (mm d^{-1}) and 850 hPa winds (m s^{-1}) from the GEOS-4 assimilated meteorological data. (c) Simulated pentad mean wet deposition (10^5kg d^{-1}) of $PM_{2.5}$ aerosols from P31 to P36. The dates corresponding to each pentad are indicated.

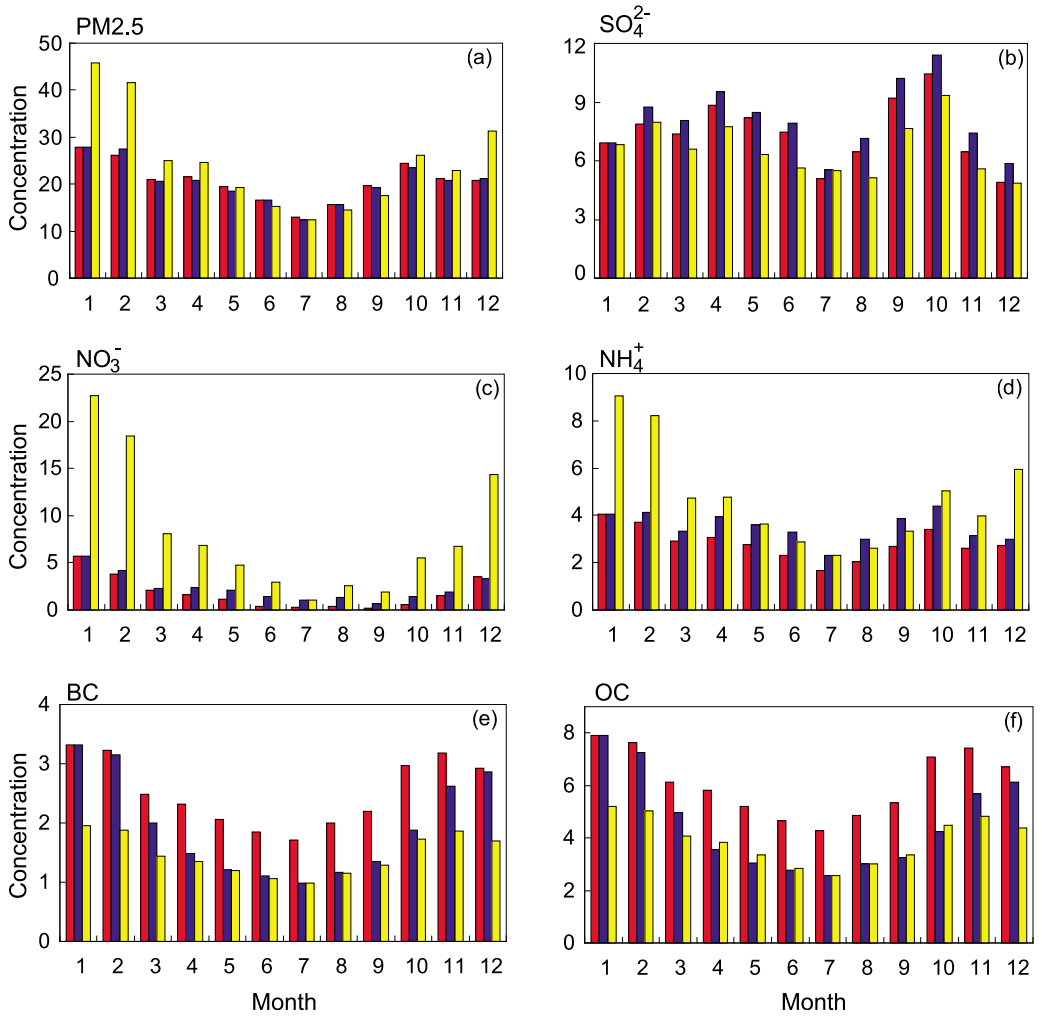

Figure 7. Comparisons of simulated surface layer aerosol concentrations (μg m^{-3}) averaged over eastern China from 2001EmAnn (blue bars), 2001EmJan (red bars), and 2001EmJul (yellow bars). (a) $PM_{2.5}$, (b) sulfate, (c) nitrate, (d) ammonium, (e) black carbon, and (f) organic carbon aerosol. $PM_{2.5}$ accounts for sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols.

winds can also be seen clearly in Figure 9b, which shows the differences in meridional wind at 925 hPa.

[30] The differences in surface air temperature (Figure 9c) between 1998 and 2002 were to a certain extent associated with the differences in winds (Figure 9b). Over entire eastern China, temperatures at 1000 hPa were cooler by 0–2 K in 1998 than in 2002 in June, as a result of the abnormal northerlies that brought cold air to eastern China or less warm air from southern China. In July and August, relative to 2002, abnormal southerlies led to warmer temperatures of up to 3 K in southeastern China and abnormal northerlies caused lower temperatures in northern China in 1998.

[31] Besides the regional differences in meteorological fields over eastern China, cross-equatorial flows were much weaker in 1998 than in 2002, which brought less clean air to China in the weak monsoon year. Figure 10 shows the time-longitude distributions of 925 hPa meridional wind averaged over 5°S–5°N for 1998 and 2002. The first channel located between 40°E–65°E was weaker in 1998 than in 2002, with a smaller maximum wind velocity in 1998. The other three channels between 65°E–140°E were apparently weaker in 1998 than in 2002. For example, the meridional wind velocity of the channel located in 120°E–135°E was about 2–4 m s^{-1} in 1998 while it was 6–10 m s^{-1} in 2002. The widths of all the channels of cross-equatorial flows were also narrower in 1998 than those in 2002.

5.2. Mass Concentrations in Weak Monsoon Year (1998) Versus Those in Strong Monsoon Year (2002)

[32] Figure 11 shows the differences in simulated surface layer concentrations of $PM_{2.5}$, BC, and sulfate between 1998 and 2002 (concentrations from 1998EmAnn minus those from 2002EmAnn) for months June, July, and August. Concentrations of all these species are generally higher in 1998 than in 2002 over eastern China, except that concentrations are lower over southeastern China in July and August of 1998. If we divide the domain of eastern China (110°E–120°E, 20°N–45°N) into northern China (110°E–120°E, 31°N–45°N) and southern China (110°E–120°E, 20°N–30°N), surface layer concentrations of $PM_{2.5}$ are predicted to increase by about 10 μg m^{-3} (or 30–60% higher relative to 2002) over northern China, and to decrease by

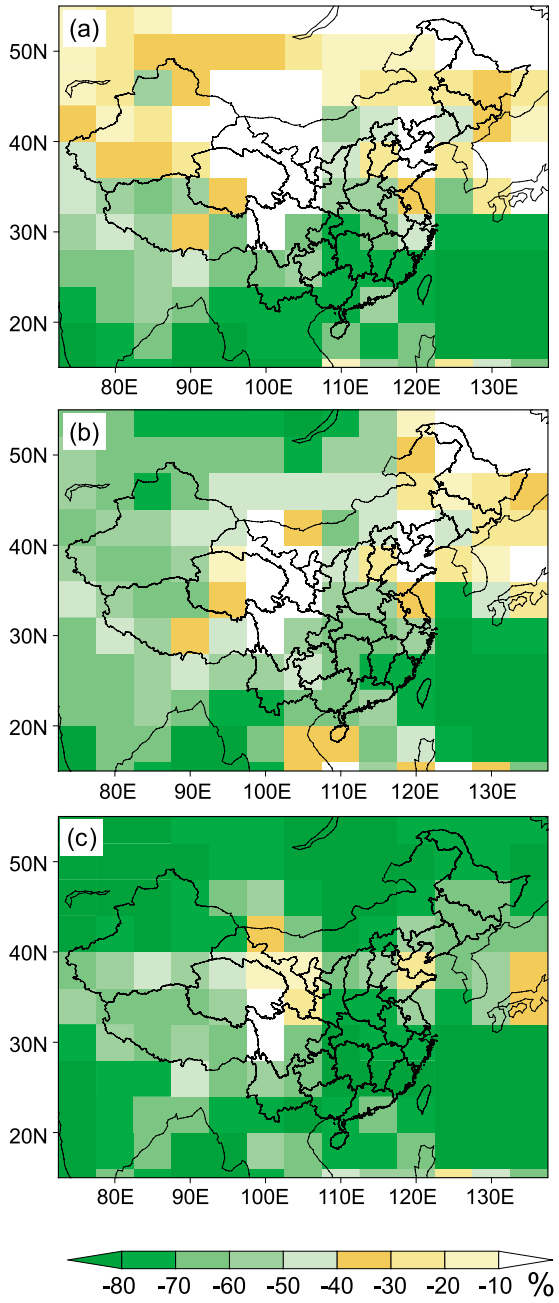

Figure 8. Reduction rates estimated by $R = ([PM_{2.5}]_{Jul} - [PM_{2.5}]_{Jan})/[PM_{2.5}]_{Jan}$ for each of simulations of (a) 2001EmAnn, (b) 2001EmJan, and (c) 2001EmJul. $[PM_{2.5}]_{Jan}$ and $[PM_{2.5}]_{Jul}$ represent monthly mean concentrations of $PM_{2.5}$ in January and July, respectively. $PM_{2.5}$ accounts for sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols.

less than 2 $\mu g\ m^{-3}$ over southern China in July and August, as summarized in Table 3. The location of maximum differences in $PM_{2.5}$ migrates northward from June to July along with the northward movement of monsoon system. With sulfate accounting for about half of $PM_{2.5}$ mass, the pattern of differences in sulfate between 1998 and 2002 is similar to that in $PM_{2.5}$. Since the strength of the monsoon can influence aerosol concentrations by altering both transport and chemical reactions, we can identify the roles of transport and chemical reactions by examining the patterns of differences in BC and sulfate, considering that BC is a chemically inert tracer. The pattern of changes in sulfate is similar to that of differences in BC, suggesting that the impact of the monsoon strength on aerosols is mostly through changing the transport and wet deposition other than chemical reactions of aerosols.

[33] Although surface layer aerosol concentrations are lower over southern China in July and August of the weak monsoon year of 1998, column burdens of $PM_{2.5}$, BC, and sulfate over entire eastern China are all higher in 1998 than in 2002 (Figure 12). The reasons for such behavior will be discussed in detail in section 5.3.

[34] Table 3 shows the differences in surface layer concentrations and column burdens of each aerosol species and $PM_{2.5}$ between 1998 and 2002 (concentrations in 1998 minus those in 2002) that are averaged over northern China and southern China. For surface layer concentrations, the behavior of each aerosol species is generally similar to that of $PM_{2.5}$, with positive changes in northern China in June–August and negative changes in southern China in July and August. It should be mentioned here that long-term measurements of $PM_{2.5}$ are not available for evaluation of predicted interannual variations of aerosols in China. *Chan and Yao* [2008] showed that measured annual mean PM_{10} concentration in Shanghai was higher by about 50% in 1998 than in 2002, which, to some extent, supported our predictions, although PM_{10} is different from $PM_{2.5}$ and the measured concentrations included the effects of differences in both meteorological fields and emissions between these two years.

5.3. Factors That Contribute to the Differences in Concentrations Between Weak and Strong Monsoon Years

[35] With the same emission inventories in simulations 1998EmAnn and 2002EmAnn, differences in concentrations of aerosols between 1998 and 2002 are a net effect of differences in transport, deposition, and chemical reactions resulting from the differences in meteorological fields between these two years. We investigate here how the differences in meteorological parameters contribute to the differences in aerosol concentrations between 1998 and 2002.

5.3.1. Precipitation and Wet Deposition

[36] With the strong rainfall associated with the Asian summer monsoon, differences in precipitation between weak and strong monsoon years are expected to have a significant effect on aerosol concentrations. Comparisons of precipitation in Figure 9a with concentrations in Figures 11 and 12 indicate that the pattern of differences in concentrations does not follow that of differences in precipitation. In southern China in June, predicted concentrations of aerosols were higher while precipitation was heavier in 1998 than in 2002. In July, a general heavier precipitation over entire eastern China led to only small reductions in surface layer concentrations in southern China in 1998 (Figure 11 and Table 3). In August, lower the less precipitation in southern China did not agree with the lower aerosol concentrations there in 1998. Furthermore, the overall larger precipitation over eastern China in 1998 did not lead to

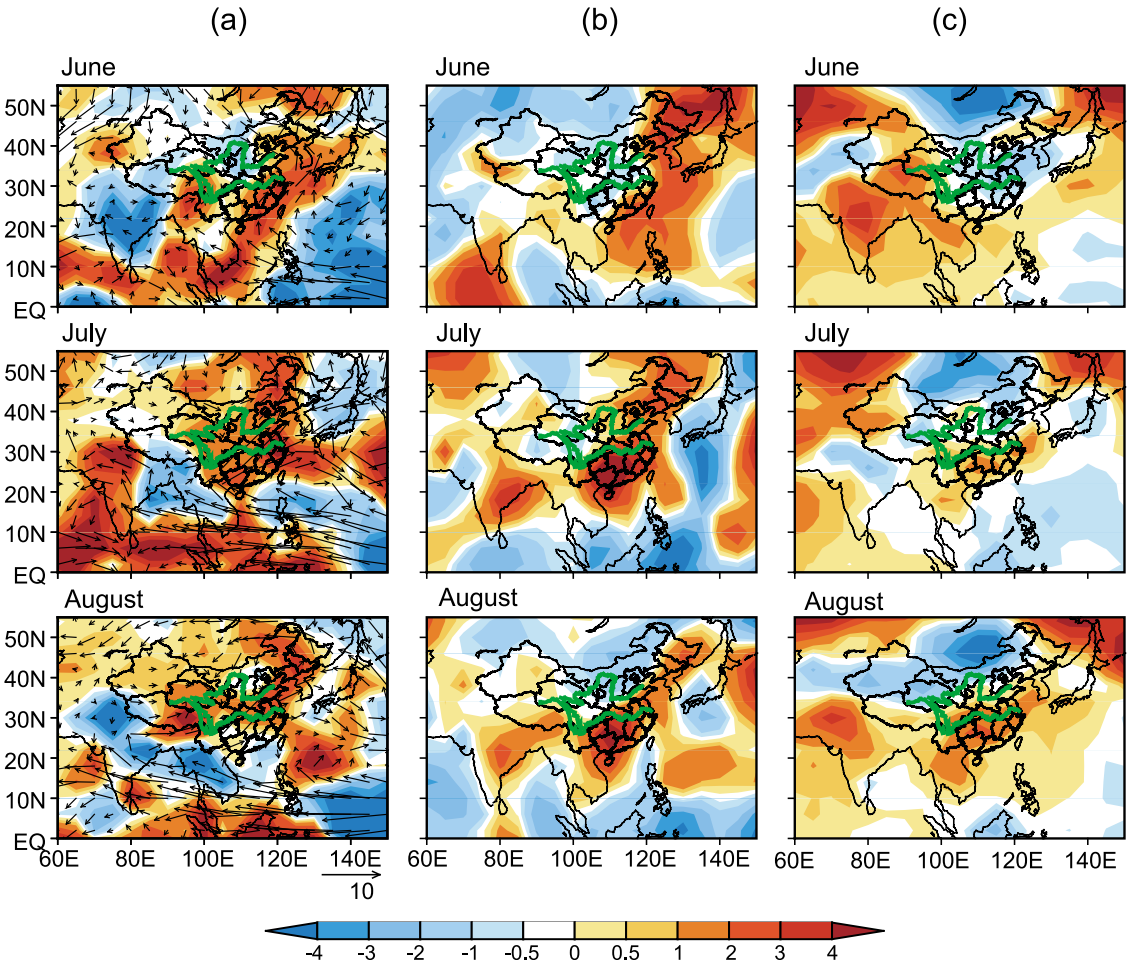

Figure 9. The differences in meteorological parameters between the weak monsoon year 1998 and the strong monsoon year 2002 (1998 minus 2002 values) for summer months June, July, and August. (a) Precipitation (mm d^{-1}) and wind at 850 hPa (m s^{-1}), (b) meridional wind at 925 hPa (m s^{-1}), and (c) temperature at 1000 hPa (K).

lower aerosol column burdens in 1998; column burdens of all aerosol species increased relative to 2002. These inconsistencies indicate that differences in precipitation are not the only factors that influence interannual variation of aerosols in eastern China.

[37] Figure 13a shows the differences in wet deposition of $PM_{2.5}$ aerosols between 1998 and 2002 (1998EmAnn – 2002EmAnn). Negative changes in wet deposition are predicted in almost all of entire eastern China in June and August and in southern China in July. In June of 1998, the negative changes in northern China were associated with the reduced precipitation in the weak monsoon year 1998 (Figure 9a), and those in southern China can be explained by the stronger convection associated with the heavier rainfall in 1998. Aerosols that were transported to the middle and upper troposphere by convection were subjected to less wet deposition than they would have been in the lower troposphere. Larger upward mass flux of $PM_{2.5}$ in 1998 than in 2002 can be seen clearly in Figure 13b (the upward flux includes contributions from both large-scale vertical transport and convective transport). In July of 1998, while the larger wet deposition in northern China agreed with the increases in rainfall in that region relative to 2002, the negative changes in wet deposition in southern China were caused by both the stronger upward transport of aerosols by convection and northward transport of aerosols associated with the stronger southerlies in southern China (Figures 9a and 9b), both of which removed aerosols from the lower troposphere of southern China. In August of 1998, the reductions in wet deposition in northern and southern China corresponded to the lighter precipitation (Figure 9c), while the reductions in southern China were enhanced by the upward and northward transport of aerosols. The reduced wet deposition of aerosols in June contributed to the predicted overall higher surface concentrations and column burdens of aerosols in 1998 than in 2002. The reduced wet deposition in southern China in July and August did not lead to increases in surface concentrations in that region, indicating the role of differences in transport.

5.3.2. Transport of Aerosols

[38] The strength of the Asian summer monsoon influences aerosol transport by the differences in cross-equatorial flows, winds from South Asia, and local winds in China. Relative to 2002, winds in the weak monsoon year 1998 contribute to the increases in aerosol concentrations in eastern China in a number of ways. First, on a large spatial

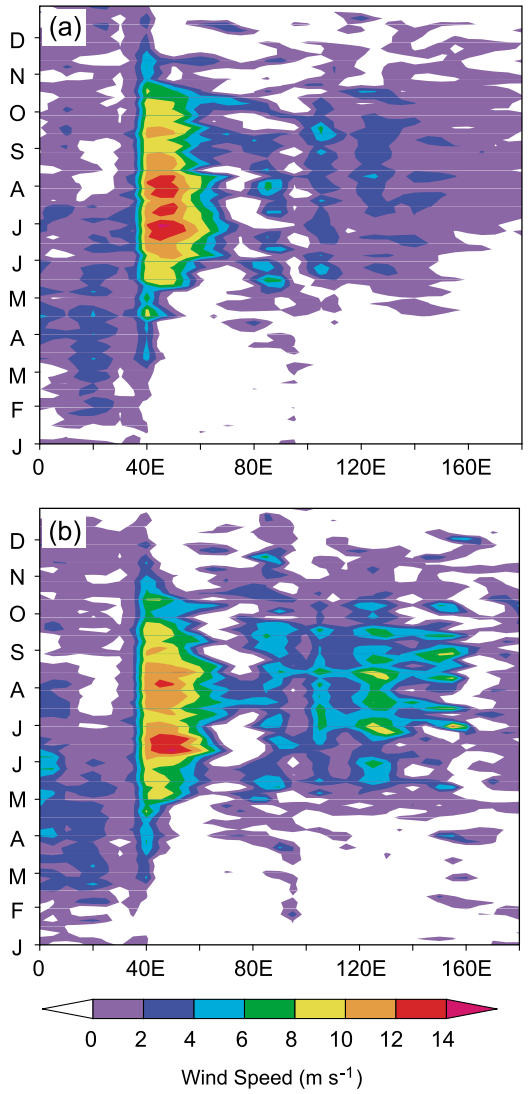

Figure 10. Time-longitude distribution of 925 hPa meridional wind (m s^{-1}) averaged over 5°S–5°N for (a) weak monsoon year 1998 and (b) strong monsoon year 2002. Meteorological fields are from the GEOS-4 assimilated meteorological data.

scale, weaker cross-equatorial flows in 1998 brought less clean air to China, contributing to the higher $PM_{2.5}$ concentrations in eastern China in 1998. Second, less precipitation led to higher concentrations of aerosols in South Asia in 1998, which increased the transport of aerosols from South Asia to China. Third, abnormal northerlies (stronger northerlies or weaker southerlies) occurred over northern China, preventing the northward transport of aerosols and leading to the build up of aerosols over eastern China. Forth, the stronger northeasterlies over the western North Pacific at 40°N in July prevented the outflow of pollutants from eastern China. Finally, stronger convection in 1998 transported aerosols to the middle and upper troposphere, leading to less wet deposition of aerosols as mentioned above. These changes in transport play a very important role in influencing the interannual variation of aerosols over eastern China.

5.3.3. Chemical Reactions

[39] Differences in meteorological fields can influence chemical reactions that lead to formation of sulfate, nitrate, and ammonium. The major factors that influence aerosol formation are temperature and relative humidity [*Dawson et al.*, 2007]. While higher temperatures can increase oxidation of SO_2 in both the gas and aqueous (cloud) phases [*Seinfeld and Pandis*, 2006], they shift aerosol thermodynamic equilibrium to gas phase and reduce concentrations of ammonium and nitrate. Temperatures over northern China were lower in 1998 than in 2002 (Figure 9c), which should result in reductions in sulfate production. However, predicted sulfate concentrations in northern China are higher in 1998 than in 2002, indicating that differences in wet deposition and transport are more important than the changes in chemical reactions in influencing sulfate concentrations as the role of a weak monsoon year is compared with that of a strong monsoon year. Although nitrate formation is sensitive to temperature and relative humidity [*Dawson et al.*, 2007], the differences in these meteorological parameters between weak and strong monsoon years do not have a large impact on nitrate formation; the differences in nitrate concentrations between 1998 and 2002 have the same qualitative changes as those in chemically inert tracer BC.

6. Seasonal and Interannual Variations of Aerosols in Eastern China in the Presence of Mineral Dust and Sea Salt

6.1. Predicted Concentrations of Dust and Sea Salt

[40] As mentioned in section 2.1, the GEOS-Chem model simulates dust aerosol in four size bins (0.1–1.0, 1.0–1.8, 1.8–3.0, and 3.0–6.0 μm dry radius), submicron sea salt, and supermicron sea salt. We focus on simulated dust in the size bin of 0.1–1.0 μm and submicron sea salt, because large particles cannot be transported far away from the emission sources to eastern China as a result of the gravitational settling. Figure 14a shows the simulated surface layer concentrations of mineral dust in the size bin of 0.1–1.0 μm in January and July of 2001 (obtained from simulation 2001EmAnn). Dust concentrations are predicted to be higher in July than in January. In July, the maximum concentrations of about 20 μg m^{-3} are predicted over the Takalamakan Desert in western China, and dust particles transported to northern China (110°E–120°E, 31°N–45°N) have concentrations of about 10 μg m^{-3}. In January, maximum dust concentrations of 6–8 μg m^{-3} are predicted over northern China. The consideration of dust can improve the comparisons of the simulated $PM_{2.5}$ concentrations with observations shown in Table 2, but the low bias in model predictions still exists. Averaged over eastern China, small dust particles (with radii of 0.1–1.0 μm) have the highest concentration of 34.6 μg m^{-3} in April (Table 4), which agree with the observed seasonal variation of dust in China [*Fairlie et al.*, 2007]. Sea salt concentrations averaged over eastern China are very small (less than 1.0 μg m^{-3}) throughout the year.

6.2. Predicted Aerosol Optical Depth

[41] Optical properties are calculated in the GEOS-Chem model for each aerosol component as a function of local

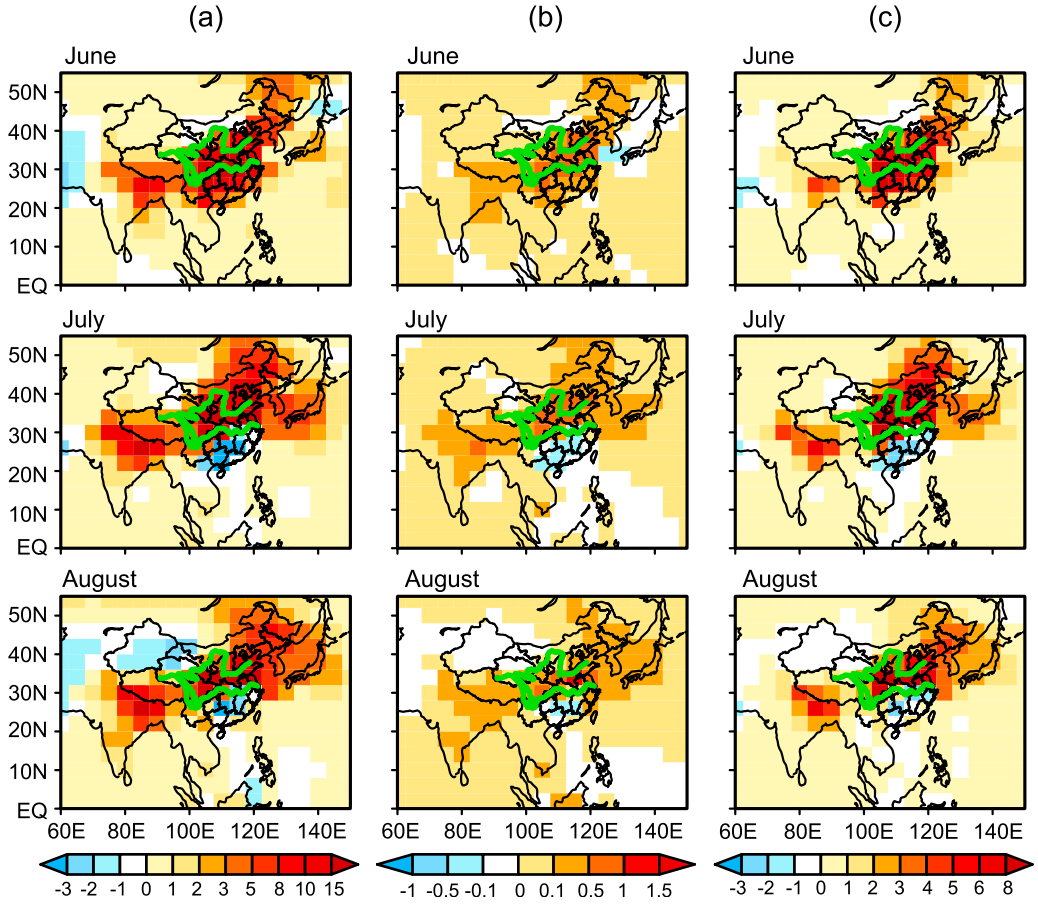

Figure 11. Differences in simulated surface layer concentrations (μg m^{-3}) of (a) $PM_{2.5}$, (b) BC, and (c) sulfate between weak monsoon year 1998 and strong monsoon year 2002 (1998 minus 2002 values) for months June, July, and August. $PM_{2.5}$ accounts for sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols.

relative humidity (RH), as described by *Martin et al.* [2003]. All aerosols are treated as externally mixed with lognormal size distributions and optical properties defined by the Global Aerosol Data Set (GADS) [*Köpke et al.*, 1997] database and by *Ginoux et al.* [2001] and *Patterson et al.* [1977] in the case of dust. Figure 14b shows the simulated optical depth of mineral dust aerosol in all size bins for January and July of 2001. Over eastern China, dust aerosol optical depth (AOD) is about 0.1 in January and can reach 0.2 in July. When we account for all aerosol species (sulfate, nitrate, ammonium, BC, OC, dust, and sea salt), aerosol optical depths over eastern China have maximum values of about 1.0 around 30°N in January and of 0.6–0.7 around 40°N in January (Figure 14c). The simulated magnitude and distribution of AOD over eastern China agree fairly well with the MODIS measurements in 2001 (Figure14d), indicating that the GEOS-Chem simulation can capture the magnitude and spatial distribution of column burdens of $PM_{2.5}$ over eastern China. It should be noted that AODs cannot be used to investigate seasonal variation of aerosol concentrations or column burdens, since AODs depend on relative humidity, which is the highest in summer in eastern China.

Table 3. Simulated Differences in Monthly and Seasonal Mean Surface Layer Concentrations and Column Burdens of Different Aerosol Species Between 1998 and 2002[a]

	1998–2002						
	Northern China			Southern China			Eastern China
Species	June	July	August	June	July	August	JJA
Surface Layer Concentrations (μg m^{-3})							
$PM_{2.5}$	8.90	12.90	10.29	7.38	−1.68	−0.48	6.22
SO_4^{2-}	4.48	6.33	4.54	3.93	−0.80	−0.39	3.01
NO_3^-	0.87	1.85	1.78	0.70	−0.30	−0.26	0.77
NH_4^+	1.96	2.88	2.19	1.63	−0.39	−0.22	1.34
BC	0.42	0.45	0.41	0.23	−0.12	<−0.01	0.23
OC	1.18	1.39	1.36	0.89	−0.07	0.38	0.86
Column Burdens (mg m^{-2})							
$PM_{2.5}$	24.51	29.41	24.69	22.50	4.61	5.99	18.62
SO_4^{2-}	14.23	17.21	14.81	12.71	1.58	2.79	10.55
NO_3^-	0.97	1.16	0.52	0.92	0.73	0.16	0.74
NH_4^+	5.34	6.51	5.19	4.90	0.90	1.23	4.013
BC	0.88	0.96	0.85	0.73	0.16	0.29	0.65
OC	3.08	3.57	3.30	3.25	1.24	1.52	2.66

[a]Values are averaged over northern China (110°E–120°E, 31°N–45°N), southern China (110°E–120°E, 20°N–30°N), or eastern China (110°E–120°E, 20°N–45°N).

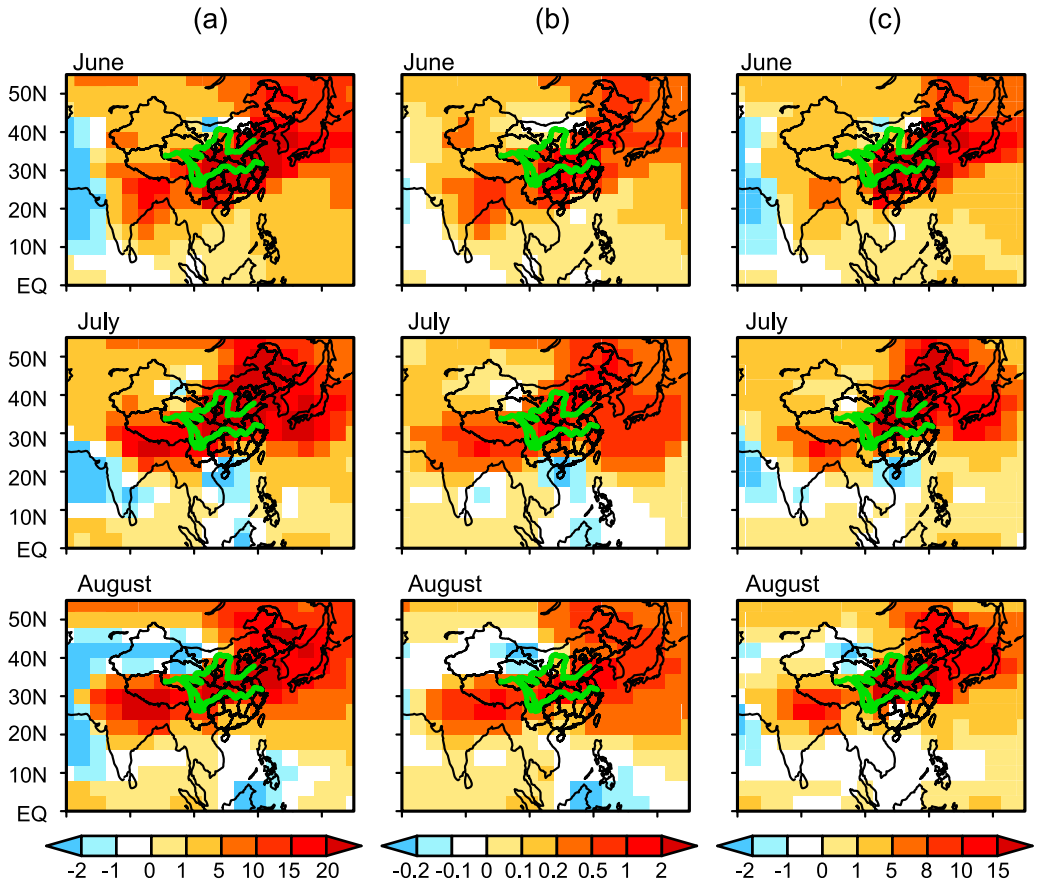

Figure 12. Differences in simulated column burden (mg m^{-2}) of (a) $PM_{2.5}$, (b) BC, and (c) sulfate between weak monsoon year 1998 and strong monsoon year 2002 (1998 minus 2002 values) for months (top) June, (middle) July, and (bottom) August. $PM_{2.5}$ accounts for sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols.

6.3. Seasonal and Interannual Variations With All Aerosols

[42] It is of interest to examine seasonal and interannual variations of $PM_{2.5}$ in eastern China in the presence of sea salt and dust. Table 4 summarizes surface layer concentrations and column burdens of $PM_{2.5}$ (sum of sulfate, nitrate, ammonium, BC, OC, mineral dust in size bin of 0.1–1.0 μm, and submicron sea salt) averaged over eastern China in January, April, July, and October of 2001, as well as those in JJA of 1998 and 2002. Because of the highest dust concentrations in April, both surface layer concentrations and column burdens of $PM_{2.5}$ show their highest value in April of 2001. However, the presence of dust and sea salt does not influence our previous conclusion on the impacts of Asian summer monsoon on seasonal variation of aerosols. Averaged over eastern China, the reduction rate $(([PM_{2.5}]_{Jul} - [PM_{2.5}]_{Jan})/[PM_{2.5}]_{Jan})$ for surface layer concentration is −50.5% with dust and sea salt, which is close to the value of −55.1% found in the absence of them.

[43] The interannual variation of aerosols is not influenced by the presence of dust and sea salt. With both of them considered, the averaged JJA $PM_{2.5}$ concentration over eastern China in the weak monsoon year 1998 is higher than that in the strong monsoon year 2002 by 7.06 μg m^{-3} (or 44.3%), and the averaged JJA column burden of $PM_{2.5}$ over eastern China in 1998 is higher than that in 2002 by 25.1 mg m^{-2} (or 73.1%), which are close to the 6.22 μg m^{-3} (or 41.7%) and 18.62 mg m^{-2} (or 65.4%), respectively, estimated in the absence of dust and sea salt.

[44] We can also examine the impact of weak/strong monsoon on aerosol optical depth. Corresponding to the differences in burdens of $PM_{2.5}$ between 1998 and 2002, AODs over eastern China can differ by 0.5–0.7 (Figure 15c), which has important implication for studying climatic effect of aerosols in China.

7. Summary and Discussions

[45] We examine the impacts of the Asian summer monsoon on seasonal and interannual variations of aerosols over eastern China using the GEOS-Chem model driven by the assimilated GEOS-4 meteorological data. Simulated surface layer $PM_{2.5}$ aerosol concentrations are generally much higher in winter than in summer over eastern China, which agree with measurements.

[46] To separate the role of changing emissions from that of monsoon in influencing seasonal variations of aerosols, we performed three simulations: (1) control simulation 2001EmAnn with monthly variations of emissions, (2) sensitivity study 2001EmJan with emissions fixed at January levels throughout the year, and (3) sensitivity study

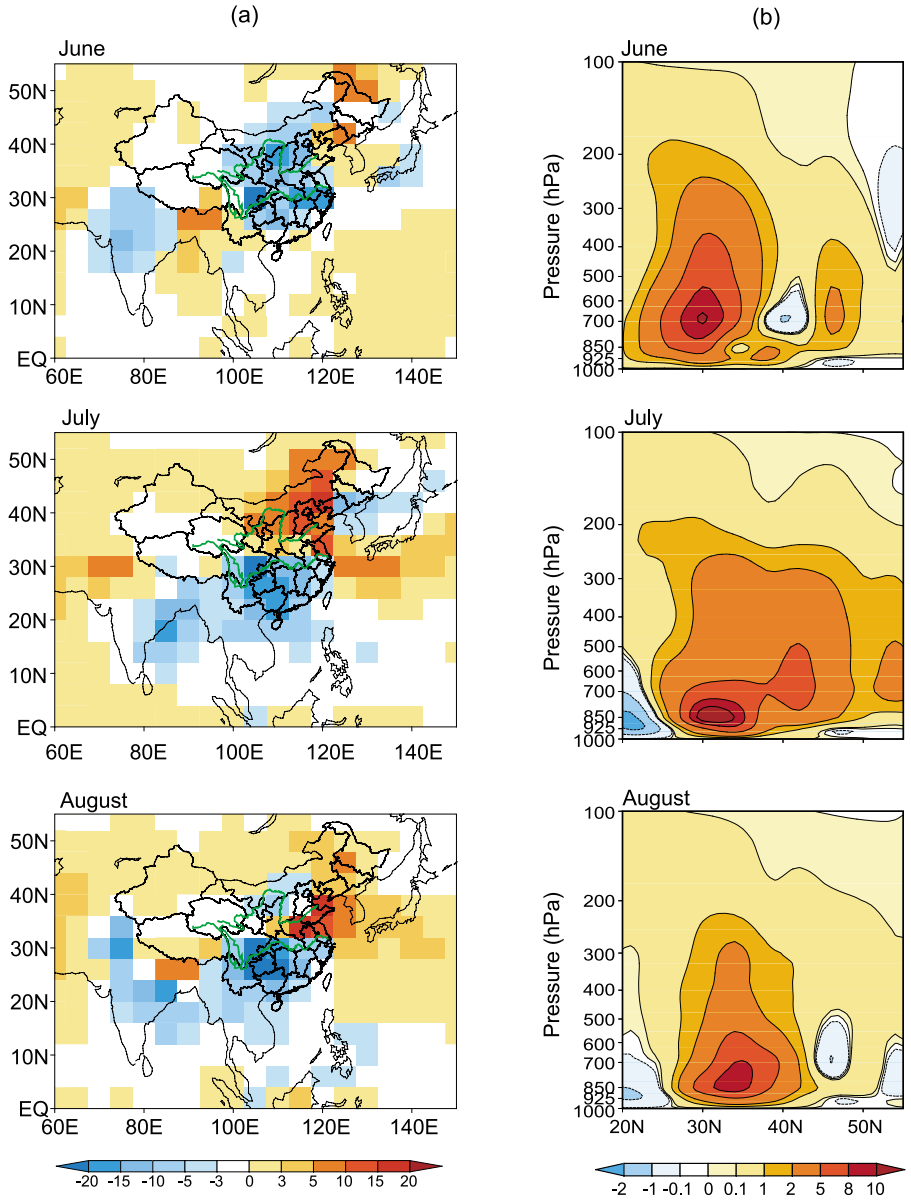

Figure 13. (a) Differences in simulated wet deposition of $PM_{2.5}$ aerosols (kg s^{-1}) between weak monsoon year 1998 and strong monsoon year 2002 (1998 minus 2002 values) for months (top) June, (middle) July, and (bottom) August. (b) Latitude-pressure plots of the differences in upward mass flux (UMF) of $PM_{2.5}$ aerosols (kg s^{-1}) between weak monsoon year 1998 and strong monsoon year 2002 (1998 minus 2002 values) for months (top) June, (middle) July, and (bottom) August. UMF values are averaged over longitude range of 110°E–120°E. $PM_{2.5}$ accounts for sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols. UMF includes contributions from both large-scale vertical transport and convective transport.

2001EmJul with emissions fixed at July levels. In all simulations, $PM_{2.5}$ concentrations over eastern China are the lowest in summer, despite the differences in emissions. Results indicate that that Asian summer monsoon is the major factor that influences the seasonal variation of $PM_{2.5}$ aerosols in eastern China. The Asian summer monsoon is predicted to reduce $PM_{2.5}$ concentration averaged over eastern China by about 50–70% in both the control run and the two sensitivity experiments, as the concentration in July is compared to that in January for each of the simulations.

[47] The cross-equatorial flows and summer monsoon rainfall play the most important roles in reducing aerosol concentrations over eastern China. Four Channels of cross-equatorial flows exist between 40°E and 135°E, which bring clean air from the Southern Hemisphere to eastern China during summer monsoon months. Correlations between cross-equatorial winds with predicted $PM_{2.5}$ concentrations in China show that different channels of cross-equatorial flows have different impacts on the spatial distributions of aerosols, with an overall effect of reducing aerosol concentrations in eastern China. Rainfall associated with sum-

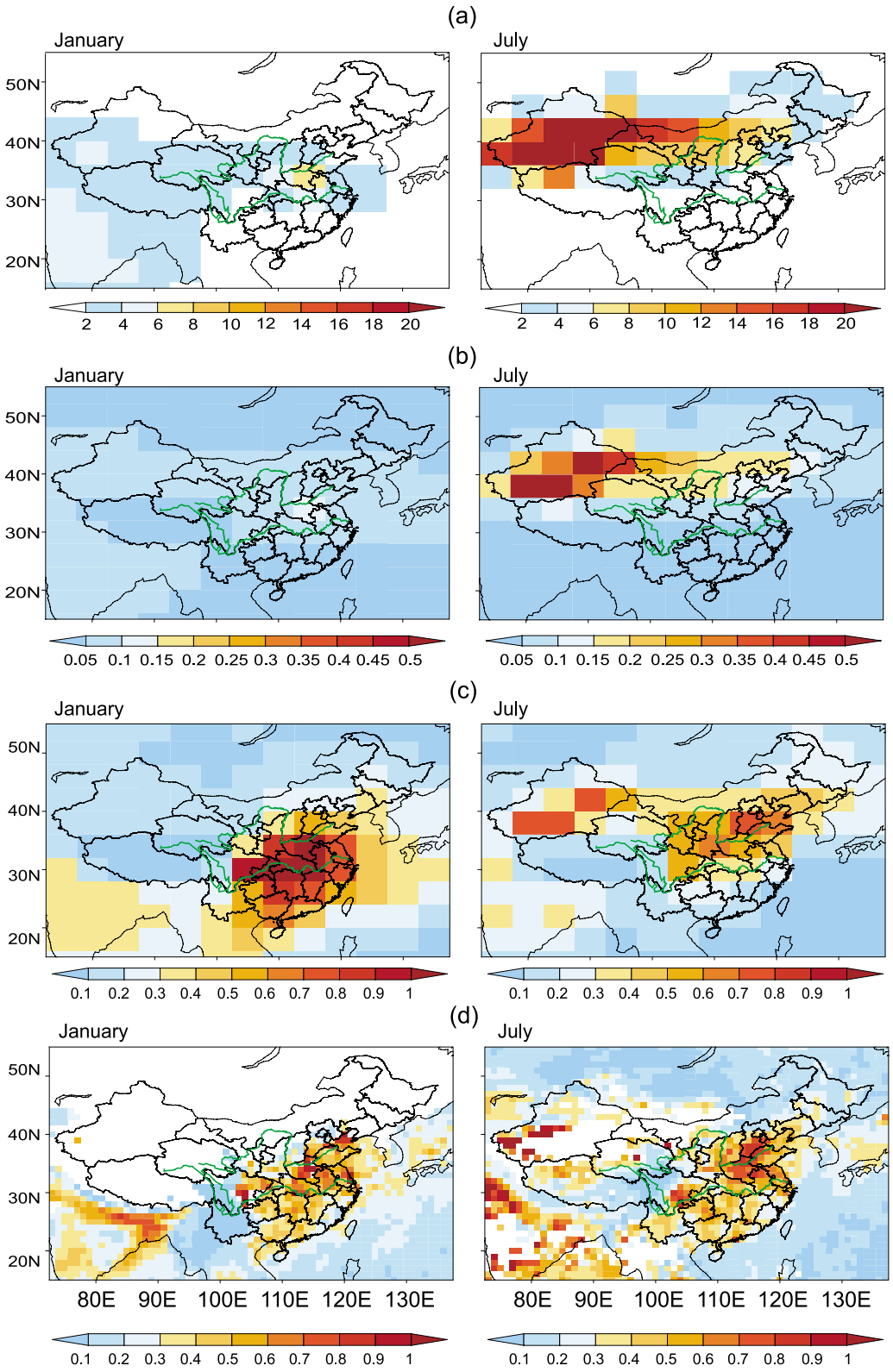

Figure 14. (a) Simulated surface layer (~1000 hPa) concentrations (μg m^{-3}) of mineral dust aerosol in the size bin of 0.1–1.0 μm dry radius, (b) simulated dust aerosol optical depth (AOD), (c) simulated AOD of all aerosols (sulfate, nitrate, ammonium, BC, OC, mineral dust, and sea salt), and (d) the MODIS measurements of AOD for (left) January and (right) July 2001.

mer monsoon leads to large wet deposition of aerosols; the reductions in $PM_{2.5}$ concentrations are sensitive to the onset and northward migration of monsoon rain belts.

[48] We assess the impacts of weak/strong monsoon years on interannual variations of aerosols over eastern China by examining the differences between aerosol concentrations simulated in 1998EmAnn and those in 2002EmAnn. With the same anthropogenic emission inventories in these two simulations, the meteorological fields of the weak summer monsoon year 1998 and the strong monsoon year 2002 are used to drive 1998EmAnn and 2002EmAnn, respectively. Model results show that both surface layer concentrations and column burdens of all the aerosol species over eastern China are generally higher in summer of 1998 relative to summer of 2002. Accounting for sulfate, nitrate, ammonium, BC, OC, mineral dust in size bin of 0.1–1.0 μm, and submicron sea salt, surface layer $PM_{2.5}$ concentration averaged over June–August over eastern China is 7.06 $\mu g\ m^{-3}$ (or 44.3%) higher in the weak monsoon year 1998 than in the strong monsoon year 2002, and column burden of $PM_{2.5}$ is 25.1 $mg\ m^{-2}$ (or 73.1%) higher in 1998 than in 2002. As a result, over eastern China, the difference in JJA aerosol optical depth between 1998 and 2002 is estimated to be up to 0.5–0.7. Analyses show the Asian monsoon strength influences interannual variations of aerosols in eastern China mostly by altering wet deposition and transport of aerosols.

[49] The Asian summer monsoon has been shown here to play major roles in influencing seasonal and interannual variations of aerosols over eastern China. Results are important for studies of air quality, climate change, and air quality-climate interactions, given that Asian summer monsoon has been weakening since the late 1970s [*Li and Zeng*, 2002, 2003; 2005; *Guo et al.*, 2003]. A number of factors contribute to the uncertainties in the model results. The first factor is uncertainties in gas and particle emission inventories that underlie any prediction of aerosol levels; such inventories are undergoing continuing improvement. A second factor relates to the consideration of biogenic secondary organic aerosols (SOAs) in the model. We assumed a 10% OC yield from biogenic isoprene in this work. Although such treatment should not have a large impact on results of this work, the SOA scheme should be updated to have more detailed treatment of biogenic SOA formation as per *Henze et al.* [2008] and *Liao et al.* [2007]. Finally, for studying interannual variations of aerosols, we focus in this work on a weak monsoon year and a strong monsoon year only; multiyear simulations using long-term assimilated meteorological data will give a more comprehensive understanding of the impact of the Asian summer monsoon and is a subject of future work.

Table 4. Simulated Average Concentrations and Column Burdens of Dust in the Size Bin of 0.1–1.0 μm, Submicron Sea Salt, and $PM_{2.5}$ Over Eastern China[a]

	2001				1998	2002	1998–2002
Species	January	April	July	October	JJA	JJA	JJA
Surface Layer Concentrations ($\mu g\ m^{-3}$)							
Dust	2.64	34.56	2.61	8.04	1.93	1.14	0.79
Sea salt	0.21	0.09	0.1	0.18	0.11	0.06	0.05
$PM_{2.5}$	30.67	55.61	15.18	31.60	22.98	15.92	7.06
Column Burdens ($mg\ m^{-2}$)							
Dust	15.87	98.13	13.30	21.30	11.90	8.30	3.60
Sea salt	0.55	0.29	0.19	0.30	0.35	0.12	0.23
$PM_{2.5}$	61.45	148.33	37.94	71.63	59.31	34.26	25.05

[a]$PM_{2.5}$ is the sum of sulfate, nitrate, ammonium, BC, OC, mineral dust in size bin of 0.1–1.0 μm, and submicron sea salt. Eastern China: 110°E–120°E, 20°N–45°N.

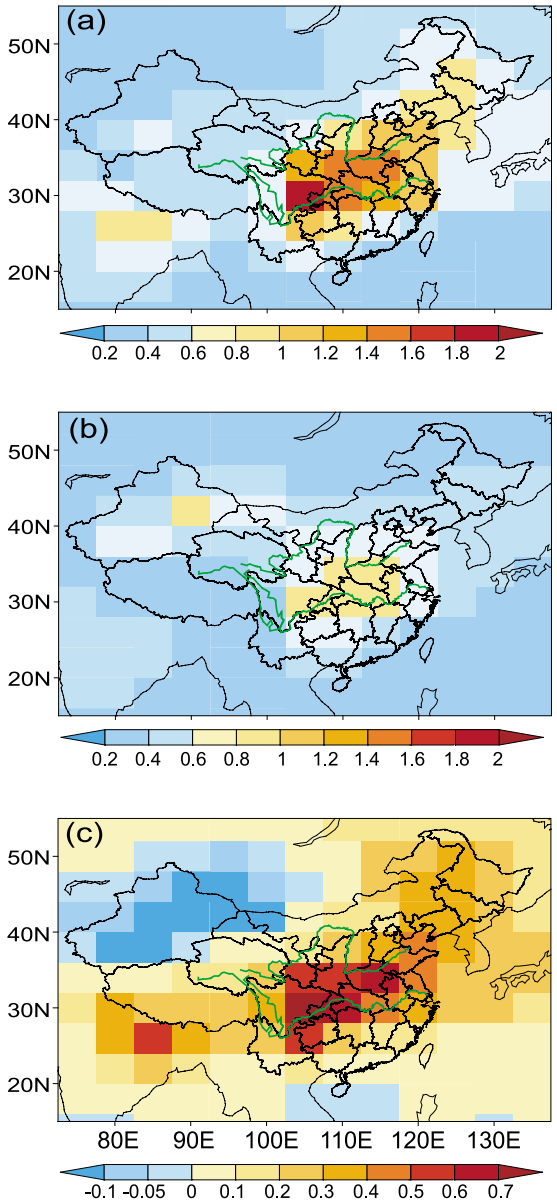

Figure 15. Simulated (a) June-July-August (JJA) mean aerosol optical depth (AOD) in 1998, (b) JJA mean AOD in 2002, and (c) differences in JJA AOD between 1998 and 2002 (1998–2002). All aerosol species (sulfate, nitrate, ammonium, BC, OC, mineral dust, and sea salt) are considered in calculations of AOD.

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References

Alexander, B., J. Savarino, C. C. W. Lee, R. J. Park, D. J. Jacob, M. H. Thiemens, Q. B. Li, and R. M. Yantosca (2005), Sulfate formation in sea-salt aerosols: Constraints from oxygen isotopes, *J. Geophys. Res.*, *110*, D10307, doi:10.1029/2004JD005659.

Aneja, V. P., J. P. Chauhan, and J. T. Walker (2000), Characterization of atmospheric ammonia emissions from swine waste storage and treatment lagoons, *J. Geophys. Res.*, *105*(D9), 11,535–11,545, doi:10.1029/2000JD900066.

Bunker, A. F. (1965), Interaction of the summer monsoon air with the Arabian Sea, paper presented at Symposium on Meteorological Results, Int. Indian Ocean Exped., Bombay.

Cao, J.-J., S.-C. Lee, K.-F. Ho, X.-Y. Zhang, S.-C. Zou, K. Fung, J. C. Chow, and J. G. Watson (2003), Characteristics of carbonaceous aerosol in Pearl River Delta Region, China during 2001 winter period, *Atmos. Environ.*, *37*, 1451–1460, doi:10.1016/S1352-2310(02)01002-6.

Cao, J.-J., S.-C. Lee, K.-F. Ho, S.-C. Zou, K. Fung, Y. Li, J. G. Watson, and J. C. Chow (2004), Spatial and seasonal variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China, *Atmos. Environ.*, *38*, 4447–4456, doi:10.1016/j.atmosenv.2004.05.016.

Cao, J. J., et al. (2007), Spatial and seasonal distributions of carbonaceous aerosols over China, *J. Geophys. Res.*, *112*, D22S11, doi:10.1029/2006JD008205.

Chan, C. K., and X. H. Yao (2008), Air pollution in mega cities in China, *Atmos. Environ.*, *42*, 1–42, doi:10.1016/j.atmosenv.2007.09.003.

Chen, L.-X., Q.-G. Zhu, H.-B. Luo, and J.-H. He (Eds.) (1991), *Eastern Asia Monsoon*, *China Meteorol.* Press, Beijing.

Chen, L.-X., H. Gao, J.-H. He, S.-Y. Tao, and Z.-H. Jin (2004), Zonal propagation of kinetic energy and convection in the South China Sea and Indian monsoon regions in boreal summer, *Sci. China, Ser. D: Earth Sci.*, *34*, 171–179.

Cooke, W. F., C. Liousse, H. Cachier, and J. Feichter (1999), Construction of a 1° × 1° fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model, *J. Geophys. Res.*, *104*(D18), 22,137–22,162, doi:10.1029/1999JD900187.

Corrigan, C. E., V. Ramanathan, and J. J. Schauer (2006), Impact of monsoon transitions on the physical and optical properties of aerosols, *J. Geophys. Res.*, *111*, D18208, doi:10.1029/2005JD006370.

Dan, M., G.-S. Zhuang, X.-X. Li, H.-R. Tao, and Y.-H. Zhuang (2004), The characteristics of carbonaceous species and their sources in PM2.5 in Beijing, *Atmos. Environ.*, *38*, 3443–3452, doi:10.1016/j.atmosenv.2004.02.052.

Dawson, J. P., P. J. Adams, and S. N. Pandis (2007), Sensitivity of PM2.5 to climate in the eastern US: A modeling case study, *Atmos. Chem. Phys.*, *7*, 4295–4309.

DeBell, L. J., K. A. Gebhart, J. L. Hand, W. C. Malm, M. L. Pitchford, B. A. Schichtel, and W. H. White (2006), Spatial and seasonal patterns and temporal variability of haze and its constituents in the United States, *Rep. IV*, Colo. State Univ., Fort Collins. (Available at http://vista.cira.colostate.edu/improve/Publications/Reports/2006/2006.htm)

Ding, Y.-H. (1992), Summer monsoon rainfalls in China, *J. Meteorol. Soc. Jpn.*, *70*, 373–396.

Duan, F. K., K. He, Y.-L. Ma, F.-M. Yang, X.-C. Yu, S. H. Cadle, T. Chan, and P. A. Mulawa (2006), Concentration and chemical characteristics of PM2.5 in Beijing, China: 2001–2002, *Sci. Total Environ.*, *355*, 264–275, doi:10.1016/j.scitotenv.2005.03.001.

Evans, M. J., and D. J. Jacob (2005), Impact of new laboratory studies of N_2O_5 hydrolysis on global model budgets of tropospheric nitrogen oxides, ozone, and OH, *Geophys. Res. Lett.*, *32*, L09813, doi:10.1029/2005GL022469.

Fairlie, T. D., D. J. Jacob, and R. J. Park (2007), The impact of transpacific transport of mineral dust in the United States, *Atmos. Environ.*, *41*, 1251–1266, doi:10.1016/j.atmosenv.2006.09.048.

Findlater, J. (1969), A major low-level air current near the western Indian Ocean during the northern summer, *Q. J. R. Meteorol. Soc.*, *95*, 91–103.

Ginoux, P., M. Chin, I. Tegen, J. M. Prospero, B. Holben, O. Dubovik, and S. Lin (2001), Sources and distributions of dust aerosols simulated with the GOCART model, *J. Geophys. Res.*, *106*(D17), 20,255–20,274, doi:10.1029/2000JD000053.

Guenther, A., et al. (1995), A global model of natural volatile organic compound emission, *J. Geophys. Res.*, *100*(D5), 8873–8892, doi:10.1029/94JD02950.

Guo, Q.-Y., J.-N. Cai, X.-M. Shao, and W.-Y. Sha (2003), Interdecadal variability of East-Asian summer monsoon and its impact on the climate of China, *Acta Geogr. Sin.*, *4*, 569–576.

He, K., F. Yang, Y. Ma, Q. Zhang, X. Yao, C.-K. Chan, S. Cadle, T. Cahn, and P. Mulawa (2001), The characteristics of PM2.5 in Beijing, *China, Atmos. Environ.*, *35*, 4959–4970, doi:10.1016/S1352-2310(01)00301-6.

Henze, D. K., J. H. Seinfeld, N. L. Ng, J. H. Kroll, T.-M. Fu, D. J. Jacob, and C. L. Heald (2008), Global modeling of secondary organic aerosol formation from aromatic hydrocarbons: High- vs. low-yield pathways, *Atmos. Chem. Phys.*, *8*, 2405–2420.

Huang, H. J., H. N. Liu, W. M. Jiang, S. H. Huang, and Y. Y. Zhang (2006), Physical and chemical characteristics and source apportionment of PM2.5 in Nanjing, *Clim. Environ. Res.*, *11–12*, 713–721, doi:10.4028/0-87849-407-3.713.

Intergovernmental Panel on Climate Change (IPCC) (2007), *Climate Change 2007: The Physical Science Basis*, Cambridge Univ. Press, Cambridge, U. K.

Jacob, D. J. (2000), Heterogeneous chemistry and tropospheric ozone, *Atmos. Environ.*, *34*, 2131–2159, doi:10.1016/S1352-2310(99)00462-8.

Joseph, P. V., and P. L. Raman (1966), Existence of low-level westerly jet streams over peninsular India, *Indian, J. Meteorol. Geophys.*, *17*, 407–410.

Köpke, P., M. Hess, I. Schult, and E. P. Shettle (1997), Global aerosol data set, report, Max-Planck-Inst. für Meteorol., Hamburg, Germany.

Lau, K. M., and K. M. Kim (2006), Observational relationships between aerosol and Asian monsoon rainfall, and circulation, *Geophys. Res. Lett.*, *33*, L21810, doi:10.1029/2006GL027546.

Lau, K.-M., G. Yang, and S. Shen (1988), Seasonal and intraseasonal climatology of summer monsoon rainfall over East Asia, *Mon. Weather Rev.*, *116*, 18–37, doi:10.1175/1520-0493(1988)116<0018:SAICOS>2.0.CO;2.

Lau, K.-M., M. K. Kim, and K. M. Kim (2006), Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau, *Clim. Dyn.*, *26*, 855–864, doi:10.1007/s00382-006-0114-z.

Li, J., and Q. Zeng (2002), A unified monsoon index, *Geophys. Res. Lett.*, *29*(8), 1274, doi:10.1029/2001GL013874.

Li, J., and Q. Zeng (2003), A new monsoon index and the geographical distribution of the global monsoons, *Adv. Atmos. Sci.*, *20*, 299–302, 10.1007/s00376-003-0016-5.

Li, J., and Q. Zeng (2005), A new monsoon index, its interannual variability and relation with monsoon precipitation, *Clim. Environ. Res.*, *10*, 351–365.

Liao, H., J. H. Seinfeld, P. J. Adams, and L. J. Mickley (2004), Global radiative forcing of coupled tropospheric ozone and aerosols in a unified general circulation model, *J. Geophys. Res.*, *109*, D16207, doi:10.1029/2003JD004456.

Liao, H., D. K. Henze, J. H. Seinfeld, S. Wu, and L. Mickley (2007), Biogenic secondary organic aerosol over the United States: Comparison of climatological simulations with observations, *J. Geophys. Res.*, *112*, D06201, doi:10.1029/2006JD007813.

Liu, H., D. J. Jacob, I. Bey, and R. M. Yantosca (2001), Constraints from 210Pb and 7Be on wet deposition and transporting a global three-dimensional chemical tracer model driven by assimilated meteorological fields, *J. Geophys. Res.*, *106*(D11), 12,109–12,128, doi:10.1029/2000JD900839.

Mari, C., D. J. Jacob, and P. Bechtold (2000), Transport and scavenging of soluble gases in a deep convective cloud, *J. Geophys. Res.*, *105*(D17), 22,255–22,267, doi:10.1029/2000JD900211.

Martin, R. V., D. J. Jacob, R. M. Yantosca, M. Chin, and P. Ginoux (2003), Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols, *J. Geophys. Res.*, *108*(D3), 4097, doi:10.1029/2002JD002622.

Menon, S., J. Hansen, L. Nazarenko, and Y. Luo (2002), Climate effects of black carbon aerosols in China and India, *Science*, *297*, 2250–2253, doi:10.1126/science.1075159.

Nenes, A., C. Pilinis, and S. N. Pandis (1998), ISORROPIA: A new thermodynamic model for multiphase multicomponent inorganic aerosols, *Aquat. Geochem.*, *4*, 123–152, doi:10.1023/A:1009604003981.

Park, R. J., D. J. Jacob, M. Chin, and R. V. Martin (2003), Sources of carbonaceous aerosols over the United States and implications for natural visibility conditions, *J. Geophys. Res.*, *108*(D12), 4355, doi:10.1029/2002JD003190.

Park, R. J., D. J. Jacob, B. D. Field, R. M. Yantosca, and M. Chin (2004), Natural and transboundary pollution influences on sulfate-nitrate-ammonium aerosols in the United States: Implications for policy, *J. Geophys. Res.*, *109*, D15204, doi:10.1029/2003JD004473.

Patterson, E. M., D. A. Gillette, and B. H. Stockton (1977), Complex index of refraction between 300 and 700 nm for Saharan aerosols, *J. Geophys. Res.*, *82*(21), 3153–3160, doi:10.1029/JC082i021p03153.

Qian, Y.-F., J. Jiang, Y. Zhang, Y.-H. Yao, and Z.-F. Xu (2004), The earliest onset area of the tropical Asian summer monsoon and its mechanisms, *Acta Meteorol. Sin*, *62*, 129–139.

Qin, Y.-J., Z.-Q. Chen, and L.-Y. Chen (1998), The characteristics of chemical compositions of atmospheric aerosols in Hong Kong, *China Environ. Sci.*, *18*, 489–493.

Ramanathan, V., C. Chung, D. Kim, T. Betge, L. Buja, J. T. Kiehl, W. M. Washington, Q. Fu, D. R. Sikka, and M. Wild (2005), Atmospheric brown clouds: Impacts on South Asian climate and hydrological cycle, *Proc. Natl. Acad. Sci. U. S. A.*, *102*, 5326–5333.

Seinfeld, J. H., and S. N. Pandis (2006), *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, 3rd ed., John Wiley, Hoboken, N. J.

Sun, Y.-L., G.-S. Zhuang, W. Ying, L.-H. Han, J.-H. Guo, M. Dan, W.-J. Zhang, Z.-F. Wang, and Z.-P. Hao (2004), The air-borne particulate pollution in Beijing—Concentration, composition, distribution and sources, *Atmos. Environ.*, *38*, 5991–6004, doi:10.1016/j.atmosenv.2004.07.009.

Tanaka, M. (1992), Intraseasonal oscillation and the onset and retreat dates of the summer monsoon over east, southeast Asia and the western Pacific region using GMS high cloud amount data, *J. Meteorol. Soc. Jpn.*, *70*, 613–629.

Tao, S., and L. Chen (1987), A review of recent research on east summer monsoon in China, in *Monsoon Meteorology*, Oxford University Press, edited by C. P. Chang and T. N. and Krishnamurti, pp. 60–92, Oxford Univ. Press, Oxford, U. K.

van Donkelaar, A., R. V. Martin, and R. J. Park (2006), Estimating ground-level PM 2.5 using aerosol optical depth determined from satellite remote sensing, *J. Geophys. Res.*, *111*, D21201, doi:10.1029/2005JD006996.

Wang, B., and Z. Fan (1999), Choice of South Asian Summer Monsoon indices, *Bull. Am. Meteorol. Soc.*, *80*, 629–638, doi:10.1175/1520-0477(1999)080<0629:COSASM>2.0.CO;2.

Wang, B., Z. Wu, J.-P. Li, J. Liu, C.-P. Chang, Y. Ding, and G.-X. Wu (2008), How to measure the strength of the East Asian Summer Monsoon? *J. Clim.*, *21*, 4449–4463, doi:10.1175/2008JCLI2183.1.

Wang, H., and R. Fu (2002), Cross-equatorial flow and seasonal cycle of precipitation over South America, *J. Clim.*, *15*, 1591–1608, doi:10.1175/1520-0442(2002)015<1591:CEFASC>2.0.CO;2.

Wang, Y., J. A. Logan, and D. J. Jacob (1998), Global simulation of tropospheric O_3-NO_x-hydrocarbon chemistry: 2. Model evaluation and global ozone budget, *J. Geophys. Res.*, *103*(D9), 10,727–10,756, doi:10.1029/98JD00157.

Wang, Y., G.-S. Zhuang, A.-H. Tang, H. Yuan, Y.-L. Sun, S. Chen, and A.-H. Zheng (2005), The ion chemistry and the source of PM2.5 aerosol in Beijing, *Atmos. Environ.*, *39*, 3771–3784, doi:10.1016/j.atmosenv.2005.03.013.

Wang, Z.-Y., and Y.-H. Ding (2008), Climatic characteristics of rainy seasons in China, *Chin. J. Atmos. Sci.*, *32*, 1–13.

Webster, P. J., V. O. Magana, T. N. Palmer, J. Shukla, R. A. Tomas, M. Yanai, and T. Yasunari (1998), Monsoons: Processes, predictability, and the prospects for prediction, *J. Geophys. Res.*, *103*(C7), 14,451–14,510, doi:10.1029/97JC02719.

Wesely, M. L. (1989), Parameterization of surface resistance to gaseous dry deposition in regional-scale numerical models, *Atmos. Environ.*, *23*, 1293–1304, doi:10.1016/0004-6981(89)90153-4.

Wu, W. S., and T. Wang (2007), On the performance of a semi-continuous PM2.5 sulphate and nitrate instrument under high loadings of particulate and sulphur dioxide, *Atmos. Environ.*, *41*, 5442–5451, doi:10.1016/j. atmosenv.2007.02.025.

Xu, J., G. A. Ding, P. Yan, and S. F. Wang (2007), Componential characteristics and sources identification of PM2.5 in Beijing, *J. Appl. Meteorol. Sci.*, *18*, 646–654.

Ye, B., X. Ji, H. Yang, X. Yao, C.-K. Chan, S. H. Cadle, T. Chan, and P. A. Mulawa (2003), Concentration and chemical composition of PM2.5 in shanghai for a 1-year period, *Atmos. Environ.*, *37*, 499–510, doi:10.1016/S1352-2310(02)00918-4.

Zhang, Q., et al. (2007), NO_x emission trends for China, 1995–2004: The view from the ground and the view from space, *J. Geophys. Res.*, *112*, D22306, doi:10.1029/2007JD008684.

Zhang, W., Y. Sun, G. Zhuang, and D. Xu (2006), Characteristics and seasonal variations of PM2.5, PM10, and TSP aerosol in Beijing, *Biomed. Environ. Sci.*, *19*, 461–468.

Zhang, X.-Y., P. Zhang, Y. Zhang, X.-J. Li, and H. Qiu (2007), The trend, seasonal cycle, and sources of tropospheric NO_2 over China during 1997–2006 based on satellite measurement, *Sci. China, Ser. D: Earth Sci.*, *50*, 1877–1884, doi:10.1007/s11430-007-0141-6.

Zhu, Q. G., J. H. He, and P. X. Wang (1986), A study of the circulation differences between East Asian and Indian summer monsoons with their interaction, *Adv. Atmos. Sci.*, *3*, 466–477, doi:10.1007/BF02657936.

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