Increases in aerosol concentrations over eastern China due to the decadal-scale weakening of the East Asian summer monsoon

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Received 19 February 2012; revised 4 April 2012; accepted 6 April 2012; published 15 May 2012.

[1] China has been experiencing increased concentrations of aerosols, commonly attributed to the large increases in emissions associated with the rapid economic development. We show by using a chemical transport model driven by the assimilated meteorological fields that the observed decadal-scale weakening of the East Asian summer monsoon also contributed to the increases in aerosols in China. We find that the simulated aerosol concentrations have strong negative correlations with the strength of the East Asian Summer monsoon. Accounting for sulfate, nitrate, ammonium, black carbon, and organic carbon aerosols, the summer surface-layer $PM_{2.5}$ concentration averaged over eastern China (110°–125°E, 20°–45°N) can be 17.7% higher in the weakest monsoon years than in the strongest monsoon years. The weakening of the East Asian Summer monsoon increases aerosol concentrations mainly by the changes in atmospheric circulation (the convergence of air pollutants) in eastern China. **Citation:** Zhu, J., H. Liao, and J. Li (2012), Increases in aerosol concentrations over eastern China due to the decadal-scale weakening of the East Asian summer monsoon, *Geophys. Res. Lett.*, *39*, L09809, doi:10.1029/2012GL051428.

1. Introduction

[2] Aerosols are major air pollutants that affect human health (U.N. Environment Programme and World Meteorological Organization, Integrated assessment of black carbon and tropospheric ozone: Summary for decision makers, 2011, http://www.unep.org/dewa/Portals/67/pdf/Black_Carbon.pdf), atmospheric visibility [*Wang et al.*, 2009], and global climate change [*Intergovernmental Panel for Climate Change* (*IPCC*), 2007]. Concentrations of aerosols are relatively high in China, which have been attributed to the increases in emissions along with the rapid economic development. A number of studies have reported that the abundances of aerosols can also be influenced by climate change based on the simulated climate change in general circulation models (GCMs) [*Liao et al.*, 2006; *Unger et al.*, 2006; *Bauer et al.*, 2007; *Dawson et al.*, 2007; *Jacob and Winner*, 2009; *Pye et al.*, 2009]. Given the uncertainties in the simulated regional climate change in GCMs, especially in the simulated changes in precipitation [*IPCC*, 2007], it has been difficult to have a robust understanding of the sign and magnitude of the changes in aerosol concentrations by regional climate change.

[3] Climate change in China is associated with the observed weakening of the East Asian summer monsoon (EASM) since 1950s [*Chang et al.*, 2000; *Ding et al.*, 2008]. A strong EASM has strong southerlies extending from southern China to northern China, a deficit of rainfall in the middle and lower reaches of the Yangtze River, and large rainfall in northern China, because the movement of the rain belts is associated with the strength of the southerlies. In contrast, in a weak EASM year, weak southerlies and a deficit of rainfall are found over northern China, and large rainfall occurs in southern China.

[4] The changes in aerosol concentrations in China are coupled with the changes in EASM. While studies in the past decades were generally focused on the impacts of aerosol direct and indirect forcing on the weakening of the Asian monsoon [*Ramanathan et al.*, 2005; *Menon et al.*, 2002; *Bollasina et al.*, 2011], few studies have quantified the impacts of the EASM on aerosol concentrations in China. Observations and modeling studies have shown that the summer monsoon influences seasonal to interannual variations of aerosol concentrations in East Asia. Ground measurements over eastern China [*Duan et al.*, 2006; *Cao et al.*, 2007; *Qu et al.*, 2010; *Ho et al.*, 2011] showed that aerosol concentrations are generally the lowest in summer because of the summer monsoon rainfall. Modeling studies also reported that the strength of the Asian summer monsoon can influence aerosol mass concentrations and optical depths over eastern Asia [*Zhang et al.*, 2010; *Yan et al.*, 2011]. However, none of the previous studies has examined the impact of the decadal-scale weakening of the EASM on aerosol concentrations over eastern China, which is essential for long-term planning of air quality and for understanding the climatic effects of aerosols.

2. Methods

2.1. Model Description and Numerical Experiments

[5] We simulate aerosols using the global chemical transport model GEOS-Chem (version 8.02.01, 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). The version of the model used here has a

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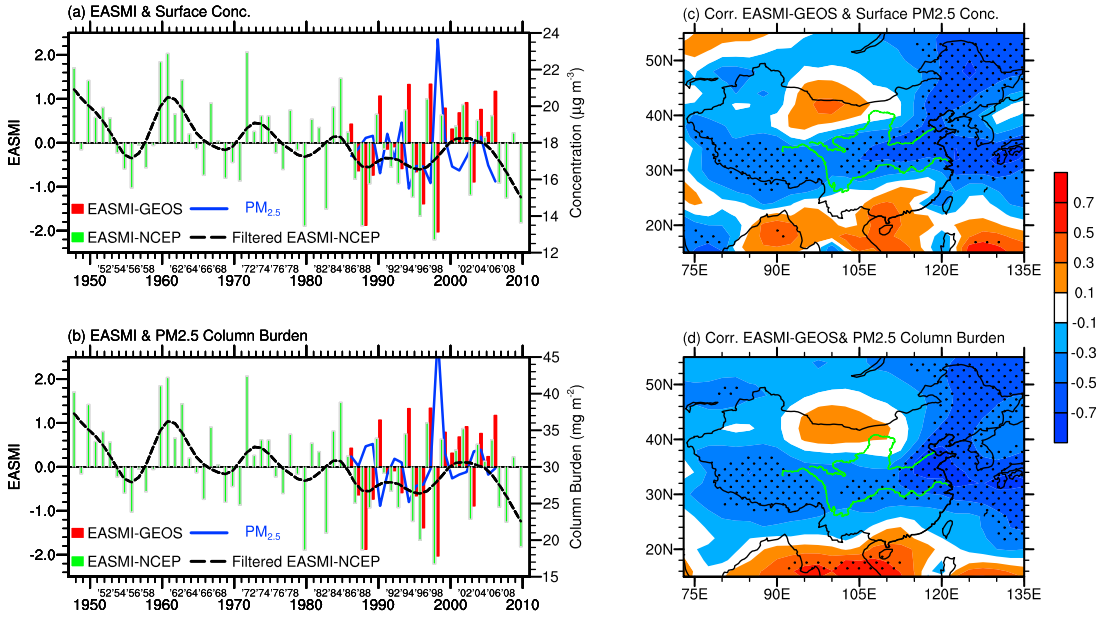

Figure 1. (a) The normalized time series of EASMI (bars, the left y-axis) and the simulated JJA surface-layer $PM_{2.5}$ concentrations (blue line, right y-axis, $\mu g\ m^{-3}$) averaged over eastern China (110°–125°E, 20°–45°N) for years of 1986–2006. The EASMI-GEOS for years 1986–2006 (red bars) are calculated with the GEOS-4 assimilated meteorological data, while the EASMI-NCEP for years 1948–2010 (green bars) are calculated using the NCEP/NCAR reanalysis data. The thick dash line is 9-year Gaussian-type filtered value of EASMI-NCEP, which represent the decadal variation of EASM. (b) The same as Figure 1a, but for the tropospheric column burdens of $PM_{2.5}$ (right y-axis, mg m^{-2}). (c) Spatial distribution of the correlation coefficients between the EASMI-GEOS and the JJA surface-layer $PM_{2.5}$ concentrations. (d) Spatial distribution of the correlation coefficients between the EASMI-GEOS and the JJA tropospheric column burdens of $PM_{2.5}$. The dotted areas areas indicate statistical significance with 95% confidence (p-value <0.05) from a two-tailed Student's t-test.

horizontal resolution of 2.5° × 2° (longitude by latitude) and 30 vertical layers from the surface to 0.01 hPa. The GEOS-Chem model includes a fully coupled treatment of tropospheric ozone-NOx-VOC chemistry and aerosols including sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), organic carbon (OC), black carbon (BC) [*Park et al.*, 2003; *Park*, 2004], mineral dust [*Fairlie et al.*, 2007], and sea salt [*Alexander et al.*, 2005].

[6] We simulate aerosol concentrations in China for years 1986–2006 driven by the GEOS-4 meteorological fields. The climate-sensitive mineral dust and sea salt aerosols are not considered in this study, because they are not major aerosol components in summer in China based on measurements [*Xuan et al.*, 2000; *Ye et al.*, 2003; *Duan et al.*, 2006]. The years of 1986–2006 are chosen for chemistry-aerosol simulation because these are the years that the GEOS-4 datasets are available. Among the GMAO meteorological products that can be used to drive the GEOS-Chem model with 2° × 2.5° resolution, the GEOS-4 datasets have the longest temporal coverage. In the simulations for 1986–2006, anthropogenic and biomass burning emissions of aerosols and aerosol precursors are fixed at the year 2005 levels. The anthropogenic emissions in Asia are taken from David Streets' emissions inventories [*Streets et al.*, 2003] and are scaled to 2005 levels (see auxiliary material, Table S1 in Text S1).[1] We fix the biomass burning emissions because no datasets are available for the whole period of 1986–2006.

2.2. The Calculation of East Asian Summer Monsoon Index (EASMI)

[7] The change in the strength of EASM is commonly represented by the East Asian summer monsoon index (EASMI). The EASMI introduced by *Li and Zeng* [2002] is used in this study to quantify the decadal-scale changes in EASM (see auxiliary material). As shown by the 9-year Gaussian-type filtered values of the EASMI (the black dashed line in Figures 1a and 1b), this smoothed time series of the EASMI changes from mostly positive values (strong monsoon years) in the years of 1948–1979 to mostly negative values (weak monsoon years) in years of 1980–2010, indicating the general trend of weakening of the EASM that has been reported by many previous studies [*Chang et al.*, 2000; *Ding et al.*, 2008].

[8] The bars in Figures 1a and 1b are the time series of the normalized EASMI, which represent the interannual variation of the strength of the EASM. We show the EASMI for years of 1986–2006 calculated with the GEOS-4 assimilated meteorological data (referred to as EASMI-GEOS) and those for years 1948–2010 calculated using the reanalyzed NCEP/NCAR datasets [*Kalnay et al.*, 1996] (referred to as EASMI-NCEP). The EASMI-GEOS agrees well with the EASMI-NCEP over 1986–2006, indicating that the GEOS-4 data has a good representation of the strength of the EASM.

3. Results

[9] Figures 1a and 1b also show simulated summertime (June-July-August, JJA) surface-layer concentrations (Figure 1a) and tropospheric column burdens (Figure 1b) of

[1]Auxiliary materials are available in the HTML. doi:10.1029/2012GL051428.

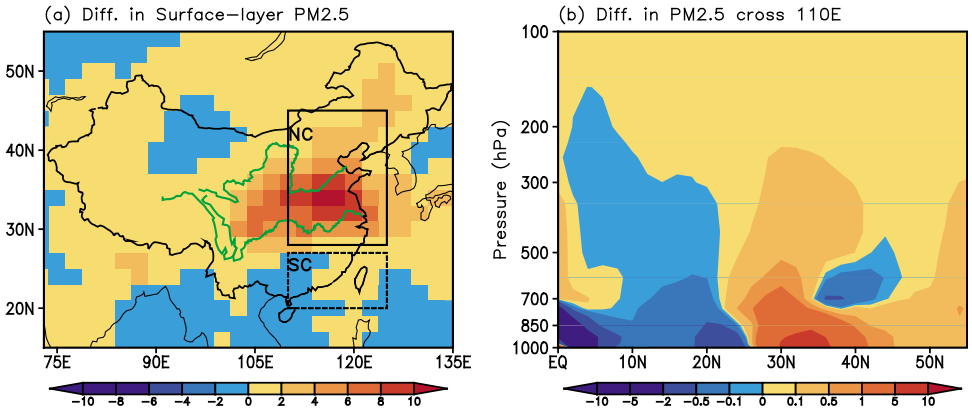

Figure 2. Composite differences of the simulated JJA surface-layer $PM_{2.5}$ concentrations between the weakest and strongest EASM years (weakest–strongest). (a) Surface layer $PM_{2.5}$ concentrations (unit: $\mu g\ m^{-3}$). (b) Pressure-latitude cross section along 110°E (unit: $\mu g\ m^{-3}$). The enclosed areas in Figure 2a define the northern China (NC, 110°–125°E, 28°–45°N) and southern China (SC, 110°–125°E, 20°–27°N) examined in this study.

$PM_{2.5}$ aerosols averaged over eastern China (110°–125°E, 20°–45°N) for 1986–2006. $PM_{2.5}$ is defined as the sum of sulfate, nitrate, ammonium, BC, and OC aerosols simulated in the GEOS-Chem model. Both the surface-layer concentrations and column burdens of $PM_{2.5}$ in summer have strong interannual variations and correlate negatively with the EASMI-GEOS; simulated aerosol concentrations/burdens are high (low) in the weak (strong) EASM years. The correlation coefficient between the surface-layer $PM_{2.5}$ concentrations and the EASMI-GEOS is −0.65, and that between the column burdens and the EASMI-GEOS is −0.55, both are statistically significant with 95% confidence level (p-value <0.05). Since we have fixed the anthropogenic and biomass burning emissions in simulations for 1986–2006, these strong negative correlations demonstrate that the monsoon strength has large impacts on JJA aerosol concentrations over eastern China.

[10] Figure 1c shows the spatial distribution of the correlation coefficients between the surface-layer $PM_{2.5}$ aerosol concentrations in JJA and the EASMI-GEOS. Large negative correlations of about −0.7 are found in central and northern China, while weak positive correlations of about 0.4 are found over the south tip of mainland China. These results indicate that a weak EASM is accompanied with high aerosol concentrations in northern China (110°–125°E, 28°–45°N) and low aerosol concentrations in southern China (110°–125°E, 20°–27°N). Similar spatial distribution and magnitude of correlation coefficients are found between the tropospheric column burdens of $PM_{2.5}$ in JJA and the EASMI-GEOS (Figure 1d).

[11] In order to quantify to what degree the weakening of EASM influences aerosol concentrations in eastern China, we examine the differences between aerosol concentrations averaged over five weakest summer monsoon years (1988, 1989, 1996, 1998, and 2003) and those averaged over five strongest summer monsoon years (1990, 1994, 1997, 2002, and 2006). These weakest (or strongest) monsoon years are selected within 1986–2006 based on the five largest negative (or positive) values of the normalized EASMI-GEOS (the red bars) as shown in Figure 1. Relative to the strongest monsoon years, the surface-layer $PM_{2.5}$ concentrations in the weakest monsoon years are higher over northern China with a maximum exceeding 10 $\mu g\ m^{-3}$, and are slightly lower by up to 1 $\mu g\ m^{-3}$ in southern China (Figure 2a). The pattern of the differences in concentrations here is in agreement with the spatial distribution of the correlation coefficients in Figure 1c. Figure 2b presents the latitude-pressure plot of the differences in $PM_{2.5}$ concentrations between the weakest monsoon years and the strongest monsoon years. Along 110°E where the maximum differences in surface-layer $PM_{2.5}$ concentrations are found, $PM_{2.5}$ concentrations from the surface to about 300 hPa are higher north of 25°N and lower south of 25°N in the weakest monsoon years than in the strongest monsoon years.

[12] The JJA concentrations of individual aerosol species (SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC) and $PM_{2.5}$ aerosols averaged over northern China and southern China are presented in Table 1 for both the weakest and strongest monsoon years. Over northern China, similar pattern of differences is found for all aerosol species (see auxiliary material, Figure S1 in Text S1); concentrations of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC are higher by 23.2%, 19.1%, 21.8%, 11.0%, and 10.4%, respectively, in the weakest monsoon years than in the strongest monsoon years (Table 1). Averaged over southern China, the differences in concentrations of all aerosol species are generally small, within the range of −0.04 to +0.03 $\mu g\ m^{-3}$ (Table 1). As a result, relative to the strongest monsoon years, $PM_{2.5}$ concentration in the weakest monsoon years is higher by 20.3% as concentrations are averaged over northern China, and by 17.7% when concentrations are averaged over the whole eastern China.

[13] The strength of the EASM influences aerosol concentrations through changes in atmospheric circulation. The difference in winds is found to be a dominant factor that leads to higher aerosol concentrations in the weakest monsoon years in northern China. Figure 3a shows the composite differences in winds at 850 hPa between the weakest and strongest monsoon years. In JJA, relative to the strongest monsoon years, anomalous northerlies are found over northern China in the weakest monsoon years, which prevent the northward transport of aerosols. Simultaneously, anomalous northeasterlies are found over the western North Pacific near 40°N, which do not favor the outflow of pollutants from northern China. In addition, the southward shift of the subtropical high in the weakest monsoon years leads

Table 1. Simulated JJA Surface-Layer Aerosol Concentrations of Different Aerosol Species Averaged Over Northern China (110°–125°E, 28°–45°N) and Southern China (110°–125°E, 20°–27°N) in the Weakest (1988, 1989, 1996, 1998, and 2003) and Strongest EASM Years (1990, 1994, 1997, 2002, and 2006)[a]

	Northern China				Southern China			
	Concentrations (μg m^{-3})				Concentrations (μg m^{-3})			
Species	Weakest	Strongest	Difference	Percentage Difference (%)	Weakest	Strongest	Difference	Percentage Difference (%)
SO_4^{2-}	10.15	8.24	+1.91	+23.2	2.88	2.85	+0.03	+0.8
NO_3^-	6.03	5.06	+0.97	+19.1	1.26	1.30	−0.04	−3.2
NH_4^+	5.45	4.48	+0.97	+21.8	1.41	1.42	−0.01	−0.7
BC	1.05	0.94	+0.11	+11.0	0.27	0.28	−0.01	−3.6
OC	1.85	1.67	+0.18	+10.4	0.65	0.66	−0.01	−1.5
$PM_{2.5}$	24.57	20.43	+4.14	+20.3	6.49	6.52	−0.03	−0.4

[a]The difference is the average over the weakest monsoon years minus that over the strongest monsoon years, and the percentage difference is (Weakest–Strongest) × 100/Strongest.

to anomalous anticyclone in the south of the middle and lower reaches of the Yangtze River (east of 105°E and south of 30°N in Figure 3a) and nearby oceans, which strengthens the northward transport of aerosols from southern China to the north of the middle and lower reaches of the Yangtze River. As a result, an anomalous convergence occurs in northern China, leading to the increases in aerosol concentrations there as shown in Figure 2a. The convergence can also be seen in Figures 3b and 3c, which show, respectively, anomalous northward and vertical transport of $PM_{2.5}$ aerosols averaged over 110°–125°E. Compared to the strongest monsoon years, increased northward mass flux of $PM_{2.5}$ aerosols exists in the lower and middle troposphere over 30°–40°N, and large increases in upward mass flux of aerosols are found over 30°–40°N in the whole troposphere, both of which confirm the anomalous convergence formed over northern China in the weakest monsoon years.

[14] The changes in rainfall associated with the strength of the EASM are expected to have a large impact on aerosol concentrations, but the impact of rainfall is not as dominant as that of the winds. Figure 3d shows the differences in wet deposition between the weakest and strongest monsoon years. Compared to the strongest monsoon years, increases in wet deposition of $PM_{2.5}$ are found in the weakest

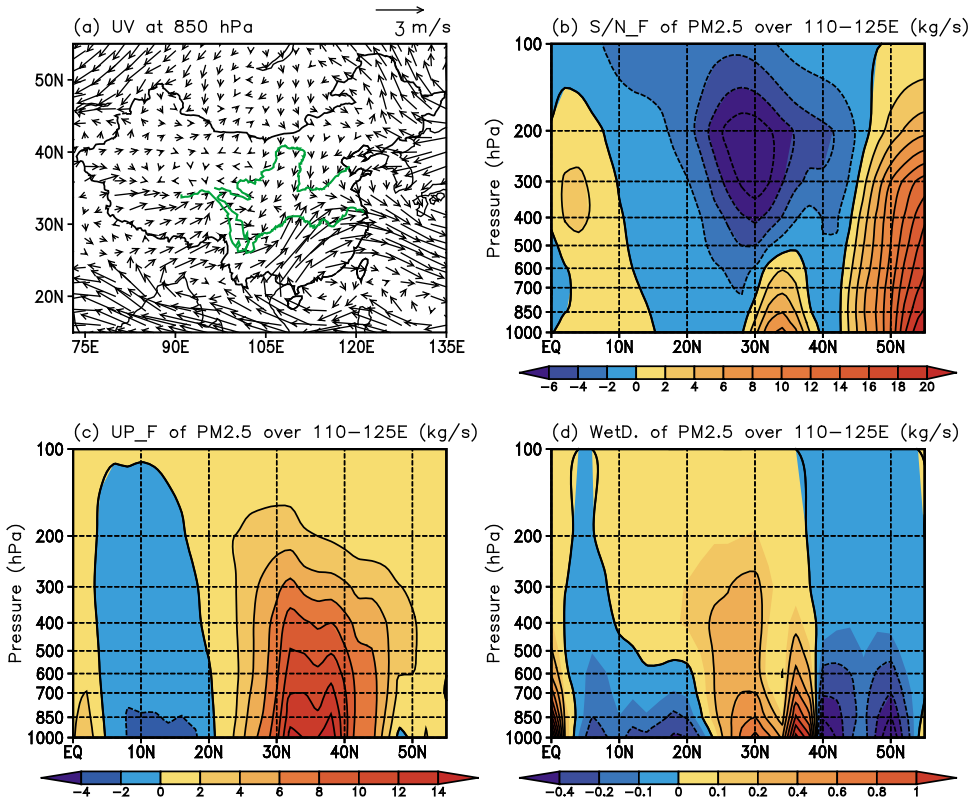

Figure 3. The composite differences in JJA parameters between the five weakest and five strongest EASM years (weakest–strongest): (a) 850 hPa wind (vector, m s^{-1}), (b) simulated northward mass flux of $PM_{2.5}$ aerosol (kg s^{-1}), (c) simulated upward mass flux of $PM_{2.5}$ aerosol (kg s^{-1}), and (d) simulated wet deposition of $PM_{2.5}$ aerosol (kg s^{-1}). Figures 3b–3d are pressure-latitude plots of the differences averaged over longitude range of 110°–125°E.

monsoon years in the whole troposphere over 25°–39°N in eastern China, as a result of the high aerosol concentrations in that region (Figure 2a) and the increases in rainfall in the lower and middle reaches of the Yangtze River (around 30°N) in the weakest monsoon years. Meanwhile, anomalously negative wet deposition of $PM_{2.5}$ is found over 40°–53°N. These changes in wet deposition are consistent with the general feature of less rainfall in northern China and increased rainfall in the middle and lower reaches of the Yangtze River (the so-called southern flooding and northern drought) in weak monsoon years relative to strong monsoon years.

[15] A sensitivity simulation is performed to quantify the role of changes in temperature associated with the variations of monsoon strength. We firstly obtain the temperature differences between the weakest and the strongest monsoon years. In the weakest monsoon years, temperatures in the lower and middle troposphere are generally lower by up to 1.2 K in northern China and higher by up to 0.6 K in southern China, as compared with the values in the strongest monsoon years (see auxiliary material, Figure S2 in Text S1). We then add the temperature differences to the temperature fields of each of the five strongest monsoon years to examine the impact of temperature changes on aerosol concentrations. The simulated aerosol concentrations with these temperature perturbations are compared with those in the 5 strongest monsoon years in Table S2 in Text S1 (see auxiliary material). The results show that the negative temperature perturbations imposed over northern China can increase $PM_{2.5}$ concentrations in the strongest monsoon years by 7.5%, mainly through the increases in sulfate, nitrate, and ammonium aerosols (see auxiliary material, Table S2 in Text S1). The SO_2 oxidation kinetics is slower at lower temperatures in our sensitivity run, which is more than offset by the decreased volatility of H_2O_2 and SO_2, increasing the in cloud aqueous-phase production of sulfate. Concentrations of sulfate show a generally negative dependence on temperature, which agree with the sensitivity analysis using the same model by *Tai et al.* [2011]. The lower temperature in northern China increases ammonium and nitrate aerosols, but leads to practically no change in SOA (or OC) because of the two competing effects on SOA concentrations (lower biogenic emissions to reduce SOA, whereas a shift in aerosol thermodynamics to favor SOA formation). As shown in Table 1, the difference in $PM_{2.5}$ concentration in northern China between the weakest and strongest monsoon years is +20.3%, in which about 7.5% (Table S2) can be explained by the temperature differences between the weakest and strongest monsoon years based on our sensitivity study. The positive temperature perturbations in southern China have an effect of reducing sulfate/nitrate/ammonium aerosols, which lead to a 9.7% reduction in $PM_{2.5}$ aerosol concentration in southern China (see auxiliary material, Table S2 in Text S1). However, the absolute changes in aerosol concentrations are small (less than 0.63 $\mu g\ m^{-3}$).

[16] We perform further simulations to compare the impacts of changing monsoon strength with those of changing anthropogenic emissions. The emissions for 1986 are simulated using the default scaling factors in the model [*van Donkelaar et al.*, 2008]. With changes in both meteorological parameters and anthropogenic emissions, $PM_{2.5}$ concentration averaged over eastern China increases from 10.6 $\mu g\ m^{-3}$ in year 1986 to 19.7 $\mu g\ m^{-3}$ in year 2006, indicating that the anthropogenic emission is still the most important factor in influencing the decadal-scale change in aerosol concentrations in China.

4. Summary and Discussion

[17] Our study concludes that the EASM strength plays an important role in determining aerosol concentrations over eastern China in summer, and the associated changes in monsoon circulation are more dominant factors than the changes in precipitation and temperature in influencing the seasonal mean aerosol concentrations. Although the strongest and weakest monsoon years mentioned above are selected form years of 1986–2006, we propose to extend our findings to estimate the decadal-scale weakening of EASM on aerosol concentrations over China based on the normalized EASMI (bars in Figure 1). Because the EASMI-GEOS agrees well with EASMI-NCEP in 1986–2006, we can use the EASMI-NCEP to represent the changes in EASM strength over 1948–2010. Since the smoothed time series of the EASMI (the black dashed line in Figures 1a and 1b) changes from mostly positive values (strong monsoon years) in the years of 1948–1979 to mostly negative values (weak monsoon years) in years of 1980–2010, we examine the EASMI-NCEP for the two periods of 1948–1979 and 1980–2010. Over 1948–1979, the averaged values of the normalized EASMI-NCEP over all 32 years, 17 years with positive values, and 15 years with negative values are +0.31, +0.97, and −0.45, respectively. Similarly, over 1980–2010, the averaged values of the normalized EASMI-NCEP over all 31 years, 15 years with positive values, and 16 years with negative values are −0.32, +0.61, and −1.19, respectively. Considering that the averaged value of the normalized EASMI-NCEP is +0.78 over the five strongest (1990, 1994, 1997, 2002, and 2006) and is −1.58 over the weakest (1988, 1989, 1996, 1998, and 2003) monsoon years that we examined above, we can conclude that the high aerosol concentrations over eastern China in recent years is partly contributed by the decadal-scale weakening of the EASM, either when we look at the averaged EASMI value over 1948–1979 (+0.31) vs. that over 1980–2010 (−0.32), or when we look at the averaged EASMI value over positive years over 1948–1979 (+0.97) vs. the averaged value over negative years over 1980–2010 (−1.19). The former comparison gives a general conclusion that the decadal-scale weakening of EASM leads to increases in aerosol concentrations in northern China, and the latter comparison indicates that aerosol concentrations in weak monsoon years in 1980–2010 can be higher than those in the strong monsoon years in 1948–1979 by about 20% based on the differences in values of EASMI and the assumption of no changes in anthropogenic emissions over 1948–2010.

[18] It should be noted that although the EASM is the dominant factor that influences summer climate in eastern China, it can be superimposed by other climate patterns such as El Niño–Southern Oscillation (ENSO) [*Feng and Hu*, 2004; *Xue and Liu*, 2008] and the annular mode in the extratropical circulation [*Nan et al.*, 2009; *Wu et al.*, 2009]. The impacts of other climate patterns on aerosol concentrations in eastern China will be a subject of our future study.

[19] **Acknowledgments.** This work was supported by the CAS Strategic Priority Research Program Grant No. XDA05100503, the National Natural Science Foundation of China under grants 40825016 and 41021004, and the special funding in atmospheric science GYHY200906020.

[20] Geoffrey Tyndall would like to thank two anonymous reviewers for their review of this paper.

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