

Impact of the Southeast Asian summer monsoon strength on the outflow of aerosols from South Asia

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Abstract. We chose a relatively weak Southeast Asian summer monsoon (SEASM) year (1998) and a relatively strong year (2002) to examine the impact of the monsoon strength on the transport of organic carbon (OC) aerosol emitted from the South Asia (75°–105° E, 10°–25° N) by using the global 3-D chemical transport model GEOS-Chem driven by the assimilated meteorological fields. Simulated surface layer concentrations and column burdens of OC indicate that OC levels are much higher in the weak SEASM year 1998 than in the strong SEASM year 2002. The sensitivity experiments with global OC emissions turned off except those over the South Asia show that OC aerosol emitted from South Asia contributes to 50-70% of OC mass over southern China and 20-50% of OC over the western North Pacific between 850 hPa and 400 hPa in 1998. The outflow of OC from the South Asia is larger in 1998 than in 2002. Three factors contribute to the larger buildup of summer time OC in the weak SEASM year of 1998. The first is the weakened summer monsoon rainfall over the Southeast Asia that leads to less wet deposition and higher OC concentrations. The second is the enhancement of deep convection in the western Indian continent and the weakened upward lifting over the western North Pacific. The last and the most important factor is the abnormal circulation in the lower and middle troposphere that contributes to the long-range transport of OC from South Asia to Southern China and the western North Pacific.

Keywords. Atmospheric composition and structure (Aerosols and particles) – History of geophysics (Atmospheric sciences) – Meteorology and atmospheric dynamics (Climatology)



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1 Introduction

South Asia is one of the areas with high aerosol concentrations, as a result of recent rapid urbanization and population growth (Chung et al., 2002; Li et al., 2005; Fu et al., 2007). Anthropogenic biomass burning has existed for thousands of years in some South Asian countries, which is a large source of air pollutants (Haberle et al., 2001). The Indian Ocean Experiment (INDOEX) (Ramanathan et al., 2001) revealed that a 3-km-thick brownish haze layer, composed of anthropogenic (accounting for up to 75% of the observed aerosol optical depth (AOD)) (Lelieveld et al., 2001) and natural aerosols, spreads over most of the tropical Indian Ocean and the Himalayan region (Ramana et al., 2004) and extends from southeastern Asia to the western Pacific (e.g., Rajeev et al., 2002).

Organic carbon (OC) aerosol is a major air pollutant that comprises a significant and sometimes major (20–90%) mass fraction of the submicron aerosols (Kanakidou et al., 2005). The average of the measured concentrations of OC in 14 major cities in China in year 2003 was 38.1 µg m⁻³ in winter and 13.8 µg m⁻³ in summer (Cao et al., 2007), which were about 20% of the averaged PM_{2.5} (aerosol particles with aerodynamic diameter less than 2.5 microns [µm]) mass in those cities and seasons. An anthropogenic source of OC in the free troposphere (FT) would have significant implications for intercontinental pollution transport and for radiative forcing of climate (Heald et al., 2005, 2006). Besides the roles of OC in air quality and climate change, OC is a chemically inert tracer that can show impacts of meteorological parameters.

Deep convection in South Asia associated with the Asian summer monsoon enhances the role of emissions in South Asia in global atmospheric chemistry. Strong vertical convections associated with summer monsoon have been shown to be important for the transport of water vapor (Randel et al., 2001; Gettelman et al., 2004; Zhan et al., 2006, 2008; Park et al., 2007), ozone (Gettelman et al., 2004; Randel and Park, 2006; Park et al., 2007, 2008), and carbon monoxide (Li et al., 2005; Fu et al., 2006; Park et al., 2007, 2008). Although Asian outflow of chemical species have been examined by a number of studies (Bey et al., 2001; Stohl, 2001; Stohl et al., 2002; Liu et al., 2003; Andreae et al., 1988; Kritz et al., 1990; Parrish et al., 1992, 2004; Berntsen et al., 1999; Yienger et al., 2000; Jacob et al., 1999; Jaffe et al., 1999, 2003; Nowak et al., 2004), most of these studies generally were focused on the intraseasonal transport in spring and considered the total outflow from both China and South Asia.

The year-to-year variation of the South Asian summer monsoon (SASM) is one of the strongest signals of the earth's climate variability. Abundant (scarce) rainfall over the Indian continent is closely associated with strong (weak) SASM (e.g., Mooley and Parthasarathy, 1984; Parthasarathy et al., 1992, 1995). Although the Indian rainfall is a good indicator of the strength of monsoon over India, it can not be used to represent the large-scale summer monsoon in the whole South Asia (Wang and Fan, 1999). The variations of summer monsoon in Indian continent and Southeast Asia (east of 85° E) are more or less independent (Lau, 1998; Wang and Fan, 1999; Li and Zeng, 2002). The atmospheric circulation and synoptic characteristics in strong summer monsoon years are quite different from those in weak summer monsoon years, which can play an important role in aerosol transport (Dawson et al., 2007). The Asian summer monsoon strength has been shown to have a large impact on aerosol concentrations over Eastern China (Zhang et al., 2010b).

Southeast Asia is located in the upstream areas of Southern China. Examining the impact of SEASM intensity on outflow of OC from the South Asia (75°-105° E, 10°-25° N) can help us to understand the interannual export of South Asian pollutants to the downwind areas and the pathways for long-range transport affected by summer monsoon strength. We address here this issue using a global three-dimensional (3-D) chemical transport model (GEOS-Chem) driven by assimilated meteorological observations. The special attention has been paid to the interannual variation of the SEASM strength in relatively weak and strong monsoon years, and then its impact on transport of South Asian pollutants to China and the western North Pacific (WNP) during summer. The structure of this paper is arranged as follows: Sect. 2 gives a description of the GEOS-Chem model, the choice of monsoon index, as well as the experimental design and data. Section 3 presents the model results for examining the different impacts of strong/weak SEASM on OC transport. Discussions and conclusions are given in Sect. 4.

2 Model description and experimental design

2.1 The GEOS-Chem model

We simulate OC concentrations using the chemical transport model GEOS-Chem (version 7.3.6 with a horizontal resolution of $4^{\circ} \times 5^{\circ}$ and 30 layers up to 0.01 hPa, see http://acmg. seas.harvard.edu/geos). The simulations are driven by the assimilated Goddard Earth Observing System GEOS-4 meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO). The GEOS-Chem model includes a fully coupled treatment of tropospheric ozone-NO_x-VOC chemistry and sulfate, nitrate, ammonium, 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 the GEOS-Chem model is described by Park et al. (2003). 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 N₂O₅ on different types of aerosols (Evans and Jacob, 2005) and irreversible absorption of NO₃, NO₂ and HO₂ on wet aerosols (Jacob, 2000).

OC is represented in the model by one hydrophobic and one hydrophilic tracer. Global missions of anthropogenic OC are taken from Cooke et al. (1999), with seasonality following Park et al. (2003). This inventory includes contributions from domestic, vehicular, and industrial combustion of various fuel types. The inventory of Cooke et al. (1999) does not include biofuels, which provide however an important source of heating in rural households and are also used in agro-industrial factories. Thus, we use a global biofuel inventory from Yevich and Logan (2003), with updates described by Park et al. (2003). Biomass burning emissions of OC are taken from the global biomass burning inventory of Duncan et al. (2003), which has monthly and interannual variability based on satellite observation of fires. We assume that 50% of OC emitted from all primary sources are hydrophobic (Cooke et al., 1999; Chung and Seinfeld, 2002), with a 1.2 day e-folding conversion from hydrophobic to hydrophilic (Cooke et al., 1999; Chin et al., 2002). We assume a 10% carbon yield of OC from terpenes (Chin et al., 2002); terpene emissions are calculated according to Guenther et al. (1995) and are dependent on vegetation type and temperature. We don't consider second organic aerosol (SOA) formation from isoprene and aromatic hydrocarbons, which may lead to an underestimate of OC concentrations over China by about $1.5 \,\mu g \, m^{-3}$ in the surface and less than $0.1 \,\mu\text{g}\,\text{m}^{-3}$ in the FT (Henze and Seinfeld, 2006; Henze et al., 2008).



Fig. 1. Simulated summer (JJAS) mean OC concentrations at 1000 hPa. (a) 1998; (b) 2002. Unit: μg m⁻³.

Fig. 2. Simulated summer (JJAS) mean tropospheric column burden OC. (a) 1998; (b) 2002. Unit: mg m⁻².

For the purpose of our study, we have replaced anthropogenic emissions (including sectors of power, industry, residential, and transportation) of OC over Asia (60° – 157.5° E, 13° S–53.5° N) by David Streets' 2006 Asian emission inventory for INTEX-B (http://www.cgrer.uiowa.edu/ EMISSION_DATA_new/index_16.html), with monthly emission profiles for each sector in China following those in the work of Zhang et al. (2007).

2.2 The strength of the SEASM and numerical experiments

The choice of an index to describe the strength of SASM is somewhat arbitrary since there are different definitions. These include indices that focus on all Indian summer rainfall (Parthasarathy et al., 1992, 1995), winds over South Asia (Webster and Yang, 1992; Goswami et al., 1999; Li and Zeng, 2002), and convection and circulation (Wang and Fan, 1999). Li and Zeng (2002) divided the broad South Asian monsoon region into two independent subsystems based on different rainfall characteristics, with the SASM sector (5°– 22.5° N, 35°–97.5° E) composed of two independent compo-

nents of the SASM1 (2.5°-20° N, 35°-70° E) and SASM2 (2.5°-20° N, 70°-110° E). The latter, more exactly speaking, is the Southeast Asia summer monsoon (SEASM) region, thus the SASM2 index here is called the SEASM index. After Li and Zeng (2002, 2003, 2005), the SEASM index measures the strength of summer (JJAS) winds over the Southeast Asia region, which has low correlation with precipitation over Indian subcontinent whereas high correlation with precipitation over Southeast Asia (east of 85° E). It can be seen from the SEASM index that 1998 and 2002 are relatively weak and strong years for the SEASM, respectively. Based on the available meteorological data, we choose these two years to analyze the impact of the SEASM strength on the transport of organic carbon (OC) aerosol emitted in South Asia. Simulations for years of 1998 and 2002 use the same OC emissions. Two simulations are performed for each of 1998 and 2002. The first simulation is the control run with global OC emissions (denoted as the simulation Emis-Globe), and the second simulation is a sensitivity run with the global OC emissions turned off except those emissions over the South Asia (75°-105° E, 10°-25° N) (including both Eastern India and Southeast Asia, denoted as the simulation

Fig. 3. Percentage changes in (a) Surface-layer OC (μ g m⁻³) and (b) Column burden of OC mg m⁻² in 1998 relative to 2002.

Fig. 4. The altitude-time cross-section of RatiosOC (colour shades) averaged over southern China $(110^{\circ}-120^{\circ} \text{ E}, 20^{\circ}-35^{\circ} \text{ N})$ for (a) 1998 and (b) 2002, and over the WNP $(125^{\circ}-150^{\circ} \text{ E}, 5^{\circ}-30^{\circ} \text{ N})$ for (c) 1998 and (d) 2002.

EmisSA). For either 1998 or 2002, the ratios of OC concentrations from the EmisSA simulation to those from the Emis-Globe simulation, denoted as RatiosOC, represent the influence of OC emitted from South Asia.

3 Model results

3.1 Surface-layer concentrations and column burdens of OC in the strong and weak SEASM years

Figure 1 shows the simulated summer (JJAS) mean surfacelayer concentrations of OC for 1998 and 2002 with global OC emissions. The spatial distributions of OC over eastern China and the Indian continent are similar in these two years. However, the magnitudes of OC are quite different between

Fig. 5. The summer (JJAS) mean RatiosOC (color shades) and wind stream from 850 hPa to 500 hPa in (a) 1998 and (b) 2002. Meteorological fields are from the GEOS-4 assimilated datasets.

the weak SEASM year (1998) and the strong SEASM year (2002). The maximum OC concentrations exceed $10 \,\mu g \,m^{-3}$ over eastern China in 1998, which nearly double those in 2002 (Fig. 1a). Meanwhile, the maximum OC concentrations located over the northeastern Indian continent are also higher

in 1998 than in 2002 (Fig. 1b). The pattern of the EmisGlobe OC column burdens in 1998 is quite different from that in 2002 (Fig. 2). Most of the China domain and northern South Asia are covered by high OC column burdens during summer of 1998 (Fig. 2a). In 2002, the pattern of OC column burdens

is similar to that of surface OC concentrations, with maximum values located in eastern China and the northeastern Indian continent (Fig. 2b). Both the surface concentrations and column burdens of OC are higher in 1998 than 2002, with the percentage changes relative to 2002 calculated as $G_{1998} = (OC_{1998} - OC_{2002})/OC_{2002} \times 100\%$ and shown in Fig. 3. Generally the large percentage increases in OC concentrations are found not only in the surface layer but also in the column burden, especially over the western North Pacific (WNP) and Indian Ocean. It should be noted that a reduction is simulated over the equatorial western North Pacific, indicating that less pollutants have been transported eastward across the equator from South Asia in 1998.

3.2 The contribution of South Asian emissions to OC in the FT over China and the WNP

The high biomass burning emissions of OC in South Asia in spring contribute to 5-50% of surface-layer OC mass in southern China, and to 40-80%, 40-60%, and 30-50% of OC concentrations at 500 hPa, respectively, over eastern China, the United States, and Europe (Zhang et al., 2010a). The EmisSA simulations with different meteorological fields of the weak (1998) and strong (2002) SEASM years are used to investigate the impacts of summer monsoon strength on OC transport from South Asia. The RatiosOC represents the contribution of South Asian OC emissions to OC concentrations over different areas. Figure 4 shows the simulated vertical distributions of RatiosOC over eastern China and Southern China in summers of 1998 and 2002. In summer (June to September), more than 50% of OC in the middle troposphere of southern China is predicted to be transported from South Asia. The high RatiosOC of up to 70% in some periods in 1998 suggest that most OC between 850 hPa and 400 hPa over southern China is not a result of local emissions (Fig. 4a). Over the WNP, in the weak SEASM year of 1998, 20-50% of OC between 850 hPa and 400 hPa is from South Asia (Fig. 4c). The impacts of South Asian OC emissions on OC over southern China and the WNP decrease largely in the strong SEASM year of 2002 (Fig. 4b, d). The differences in RatiosOC are mainly found between 850 hPa and 500 hPa during June-August.

The horizontal distributions of RatiosOC at different vertical layers are presented for 1998 and 2002 in Fig. 5. The patterns of RatiosOC at each layer in 1998 are quite different from those in 2002, indicating again the differences in OC transport in the weak and strong summer monsoon years. At 850 hPa and in 1998, RatiosOC of exceeding 30% are predicted over a large area covering from South Asia to southern China and the WNP (Fig. 5a). With increasing altitude, higher RatiosOC of up to 40–60% appear over eastern China from 700 to 500 hPa (see Fig. 5a), and the RatiosOC of 30–40% are predicted to extend eastward and southward to the WNP. In the strong SEASM year of 2002, the RatiosOC patterns in different layers are similar to each other;

Fig. 6. The differences in summer (JJAS) mean RatiosOC (color shades) and wind stream between 1998 and 2002 (the former minus latter) at (**a**) 850 hPa; (**b**) 700 hPa; (**c**) 600 hPa; (**d**) 500 hPa. Meteorological fields are from the GEOS-4 assimilated datasets.

Fig. 7. The differences in summer (JJAS) mean precipitation (mm/day) and 850 hPa winds (m/s) of GEOS4 between 1998 and 2002 (the former minus latter). Meteorological fields are from the GEOS-4 assimilated datasets.

the contributions of South Asian OC emissions to the eastward transport are less than 20% (Fig. 5b), and high RatiosOC are mainly located over South Asia without much eastward movement. The differences in RatiosOC between 1998 and 2002 (the former minus latter) are shown in Fig. 6, which represent the different contributions of South Asian OC emissions during summer. The large differences in RatiosOC are located over South Asia, tropical WNP and central WNP from 850 hPa to 500 hPa, and RatiosOC in 1998 are about 25% higher than those in 2002. Lower RatiosOC are predicted in 1998 over the Bay of Bengal (BOB), Indo-China Peninsula (ICP), and South China Sea (SCS), suggesting that a larger fraction of OC from local emissions has been transported to the downstream areas in 1998. Table 1 shows the difference in OC budget over southern China ($110^{\circ} \sim 120^{\circ}$ E, $20^{\circ} \sim 35^{\circ}$ N) in simulations EmisSA1998 and EmisSA2002. At the eastern and southern boundaries of Southern China, the OC mass fluxes are much lager in 1998 than in 2002, indicating that more OC aerosol has been transported from South Asia to southern China. The budget of fluxes shows that a larger accumulated mass of OC of 0.14 kg s^{-1} is simulated in 1998 than in 2002.

3.3 Differences in meteorological fields that contribute to the differences in OC transport

It is well known that wet deposition is a major sink for all aerosol species, so precipitation is expected to have a significant effect on aerosol concentrations. The differences in precipitation between the weak and strong SEASM years (1998 minus 2002) indicate that the rainfall of summer monsoon over the broad South Asian monsoon region is weaker in 1998 than 2002, especially in the northeastern Indian continent, BOB and the ICP (see Fig. 7). Meanwhile, relatively weaker westerlies exist in 1998 than in 2002 over the South Asia and the WNP domain, which contribute to the less transportation to the WNP. The insufficient rainfall over the broad South Asian monsoon region in the weak SEASM year results in less wet deposition of OC, which increases OC concentrations in South Asia (Fig. 8). Meanwhile, the stronger deep convection over the Indian continent in 1998 enhances the frequent lifting of OC from the surface to the FT (Fig. 9). As a result, the weak rain belts and the enhanced upward circulation over South Asia are in favor of increasing RatiosOC over China and the WNP in summer of 1998.

The seasonal mean wind streams of 1998 show that the surface-layer prevailing southeasterlies cover the northeastern Indian continent, BOB, ICP, and SCS. The stronger western Pacific subtropical high (WPSH) located to the south is in favor of transporting OC aerosol from South Asia to southern China and the WNP at 850 hPa and 700 hPa (Fig. 5a). The westward shifting WPSH can be seen at 25° N in the WNP from 700 hPa to 500 hPa, which leads to OC transport from southern China to the WNP, and then toward low latitudes (Fig. 5a). The WPSH also enhances the downward movement that weakens the convection and results in the transport of OC from the middle troposphere to the surface layer. In 2002, a SCS-WNP monsoon trough (Intertropical Convergence Zone, ITCZ) extends in the northwest-southeast direction from southern China to the WNP (Fig. 5b), and the WPSH locates at higher latitudes compared to its location in 1998. These features in circulation block the northward flows from the SCS to China and weaken the cross-equatorial flows over the WNP, which result in less northward transport of OC. Two lows located in the northeastern Indian continent and southern China bwteen 700 hPa and 500 hPa also prevent the northward and eastward transport of OC from South Asia to the downstream areas (Fig. 5b). As a result, OC aerosols are difficult to diffuse in the saddle low. At the same time, the WPSH extends westward and reaches southern China at 600 hPa and 500 hPa; the southeasterlies in the brim of the WPSH also restrain the northeastward transport of OC.

Figure 6 also shows the differences in seasonal mean atmospheric circulation between 1998 and 2002 obtained from the GEOS-4 meteorological data. The difference in pattern of atmospheric circulation agrees well with that of RatiosOC, suggesting that summer monsoon strength plays an important role in outflow of OC from South Asia. With less OC wet deposition in South Asia in 1998, the monsoonal flow is in favor of transporting more South Asian pollutants toward the northeast, during that period the pollutants may also subject to frequent lifting into the higher altitudes by the enhanced deep convection (Fig. 9). In the middle troposphere, the Tibetan anticyclone (South Asian High) is much stronger in 1998 and more OC aerosol can be transported to China and the Pacific by the westerlies. Thus, an abnormal anticyclone (A) near 20° N covers most of the broad South Asian monsoon region and the WNP, resulting in the outflow of OC firstly toward southern China and the mid-latitude of the WNP, and then toward the equator. The convection over the WNP in 1998 is much weaker than that in 2002 because of

Table 1. Summer (JJAS average) OC column burden, wet deposition, and mass fluxes ($1000 \sim 500$ hPa) in EmisSA1998 and EmisSA2002 over southern China ($110^{\circ} \sim 120^{\circ}$ E, $20^{\circ} \sim 35^{\circ}$ N).

	$\begin{array}{c} \text{Column} \\ (\text{mg}\text{m}^{-2}) \end{array}$	Wet deposition (kg s ⁻¹)	EW flux (110° E, 20° \sim 35° N; kg s ⁻¹)	EW flux $(120^{\circ} \text{ E}, 20^{\circ} \sim 35^{\circ} \text{ N}; \text{kg s}^{-1})$	NS flux $(20^{\circ} \text{ N}, 110^{\circ} \sim 120^{\circ} \text{ E}; \text{kg s}^{-1})$	NS flux $(35^{\circ} \text{ N}, 110^{\circ} \sim 120^{\circ} \text{ E}; \text{ kg s}^{-1})$
1998	0.81	0.58	3.40	1.95	0.86	1.25
2002	0.36	0.32	0.81	0.28	0.57	0.18
1998-2002	0.45	0.26	2.59	1.67	0.29	1.07

Fig. 8. The differences in simulated summer (JJAS) mean wet deposition of OC between 1998 and 2002 (the former minus latter). Unit: kg s⁻¹.

the monsoon trough (ITZC, see Fig. 9), which increases RatiosOC in the middle troposphere. Therefore, the maximum differences in RatiosOC are predicted in the southeast of the abnormal cyclone in the low latitudes of the WNP. Furthermore, abnormal cyclone (C) and anti-cyclone (A) exist over the central and northeast WNP, respectively. They couple with the abnormal anti-cyclone (A) near 20° N as a reverse "S". Such "A-C-A" pattern favors the long-distance transport of OC from South Asia, which has important implications for the export of Asian pollutants to the Pacific.

4 Conclusion and discussion

The simulated RatiosOC in a relatively weak SEASM year (1998) and a relatively strong SEASM year (2002) show that South Asian OC emissions have different contributions to OC in China and the WNP as a result of the difference in summer monsoon strength. The major factors that lead to the differences in OC transport and the higher summer time (July and August) OC buildup in southern China and the WNP in the weak SEASM year 1998 are as follows: 1) the less sum-

Fig. 9. The altitude-longitude cross-section of the differences in summer (JJAS) mean vertical circulation between 1998 and 2002 (the former minus latter). Values are averaged over latitude range of 10° – 20° N. Color Shades denote the upward movement. Meteorological fields are from the GEOS-4 assimilated datasets.

mer monsoon rainfall leads to less wet deposition and higher OC concentrations in South Asia, which can be transported to downwind areas; 2) the enhancement of deep convection in the Indian continent increases the upward lifting of OC, while the weakened convection over the WNP has an effect of transporting OC from the middle troposphere to the surface; 3) the abnormal winds (the westerlies above 850 hPa) are the most important factors that enhance the long-range transport of OC from South Asia to China and the WNP (see Fig. 6). It should be noted that although precipitation and wet depositions are larger over southern China and most of the WNP in 1998, the RatiosOC are still higher in 1998 than in 2002, suggesting that the transport of pollutants plays a very important role in influencing air quality in China.

We use the same emissions in all the numerical experiments, which allow us to detect the impacts of the strength of the SEASM. In reality, emissions in South Asia have interannual variations, which may contribute to the interannual variations in OC concentrations. Furthermore, the differences in meteorological fields can also affect biogenic OC emissions, which should be considered in subsequent studies. Although simulated seasonal variation of OC agrees with ground measurements in a number of cities over eastern China, long-term measurements of OC are needed for evaluating the simulated interannual variation of OC.

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