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Seasonal Variations of Aerosols in Pakistan: Contributions of Domestic Anthropogenic Emissions and Transboundary Transport

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ABSTRACT

Air pollution has become a serious challenge for developing countries like Pakistan. Very scarce information is available regarding pollution levels in this geographic region. This study presents the first modelling work to simulate the spatial distribution and temporal variation of aerosol concentrations over Pakistan by using the Weather Research and Forecasting Model coupled with chemistry (WRF-Chem). Simulated aerosols species include sulfate, nitrate, ammonium, organic carbon, black carbon, and $PM_{2.5}$ (particles with a diameter of 2.5 μm or less), which are evaluated against ground-based observations and satellite measurements. In year 2006, simulated $PM_{2.5}$ concentrations averaged over northeastern Pakistan (71–74.5°E, 28–34°N) are 55, 48.5, 31.5, and 98 μg/m^3 in January, April, July, and October, respectively. The simulated highest $PM_{2.5}$ concentration in October results from the relatively low temperatures that favor nitrate formation as well as the lowest precipitation that leads to the smallest wet deposition of all aerosol species. The simulated lowest concentration of $PM_{2.5}$ in July can be attributed to the largest precipitation associated with the South Asian summer monsoon. Sensitivity studies show that transboundary transport contributes to $PM_{2.5}$ aerosol levels in northeastern Pakistan by 10–20% in January and April and by 10–40% in July and October of year 2006. Wind over India and Pakistan is found to be the major meteorological parameter that determines the transboundary aerosol transport.

Keywords: WRF-Chem; Aerosols in Pakistan; Seasonal variations.

INTRODUCTION

Aerosols are important air pollutants that have adverse health impacts, lead to reductions in visibility and changes in climate (Intergovernmental Panel on Climate Change (IPCC), 2013). Over the past two decades, South Asian countries have undergone rapid urbanization and industrialization which dramatically increase emissions of air pollutants (Gurjar *et al.*, 2008). The health impacts from regional phenomena such as the Asian Brown Cloud (ABC) and intense winter fog episodes have made the South Asian region very important. Concentrations of major atmospheric aerosol species, including sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), black carbon (BC), and organic carbon (OC), are especially high in eastern Pakistan. Furthermore, lack of efficient monitoring and control measures of air pollution has made Pakistani cities more vulnerable as compared to the developed world and even neighboring countries. Towards filling these gaps, air quality modeling is essential and can be used, for example, to understand the emission sources, concentrations, and seasonal variations of aerosols in Pakistan.

Pakistan is located in South Asia, sharing borders with Afghanistan and Iran in the west, China in the north, the Arabian Sea in the south, and India in the east. There exist several studies about the observed aerosols in Pakistan. Husain *et al.* (2007) reported that in Lahore, the second largest city in Pakistan, the seasonal mean concentration of BC was 21.7 μg/m^3 in winter of 2005 to early 2006. Dutkiewicz *et al.* (2009) reported that the average concentration of BC in Karachi, the largest city in Pakistan, was about 10 μg/m^3 in winter and 2 μg/m^3 in summer in the years of 2006–2007. Lodhi *et al.* (2009) reported that $PM_{2.5}$ concentrations at Lahore were in the range of 53–

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476 μg/m^3 from November 2005 to December 2007. Biswas *et al.* (2008) reported average $PM_{2.5}$ concentration of 209 μg/m^3 at Lahore from December 2005 to February 2006. Measurements by the Environmental Protection Agency (EPA) of Pakistan reported that the average $PM_{2.5}$ concentrations over 2005–2010 in cities of Islamabad, Lahore, Karachi, Peshawar, Quetta were 73.0, 121.8, 53.2, 70.2, and 47.1 μg/m^3, respectively (Economic Survey, 2009–10). Measurements showed that nitrate, sulfate, and ammonium were the largest components that contributed to $PM_{2.5}$ at Lahore (Biswas *et al.*, 2008; Lodhi *et al.*, 2009; Stone *et al.*, 2010) and sulfate was the dominant species in $PM_{2.5}$ in Karachi (Ghauri *et al.*, 1994; Mansha *et al.*, 2012). These observational studies, however, were quite limited in terms of chemical species and spatial coverage of aerosols.

Another issue that is associated with the aerosol levels in Pakistan is the transboundary aerosol transport. Some observational analyses have demonstrated the importance of transport of aerosols from North India (Begum *et al.*, 2011). Indian coal based power plants are major sources of transboundary air pollution that influence northeastern Pakistan (Ghauri, 2010). Despite of the importance of transboundary transport, none of the previous studies has quantified the contributions to aerosol concentrations in Pakistan by domestic anthropogenic emissions and transboundary transport.

Seasonal variations of aerosols are driven by seasonal variations in both emissions and meteorological conditions. Pakistan has four seasons: cool and dry winter from December through February, hot and dry spring from March through May, the summer rainy season (or monsoon period) from June through September, and the retreating monsoon period of October and November. Adhikary *et al.* (2007) reported that aerosol concentrations in South Asia showed minimum values in the monsoon period (June–September) and maximum levels in the post monsoon season (October–November).

The aim of this study is to understand the seasonal variations of aerosols and the impacts of transboundary transport on aerosol levels in Pakistan. The Weather Research and Forecasting Model coupled with Chemistry (WRF-Chem) (Grell *et al.*, 2005) is employed to simulate aerosol concentrations over Pakistan during the months of January, April, July, and October of year 2006. The paper is organized as follows. The WRF-Chem model setup, emissions, numerical experiments as well as model evaluation on meteorological parameters, aerosol concentrations, and aerosol optical depth are described in the following sections. Simulated seasonal variations of aerosols over Pakistan and evaluations of simulated aerosol concentrations are then presented. Finally, simulated impacts of transboundary transport on aerosol levels in Pakistan are presented.

MODEL DESCIPTION AND NUMERICAL SIMULATIONS

The WRF-Chem Model

This study uses the version 3.2.1 of the WRF-Chem, developed by National Oceanic and Atmospheric Administration (NOAA) and National Center for Atmospheric Research (NCAR) (http://ruc.noaa.gov/wrf/WG11/). The model domain is defined by a Lambert projection centered at 30°N and 70°E and covers Pakistan at 30 km spatial resolution as shown in Fig. 1. The WRF-Chem

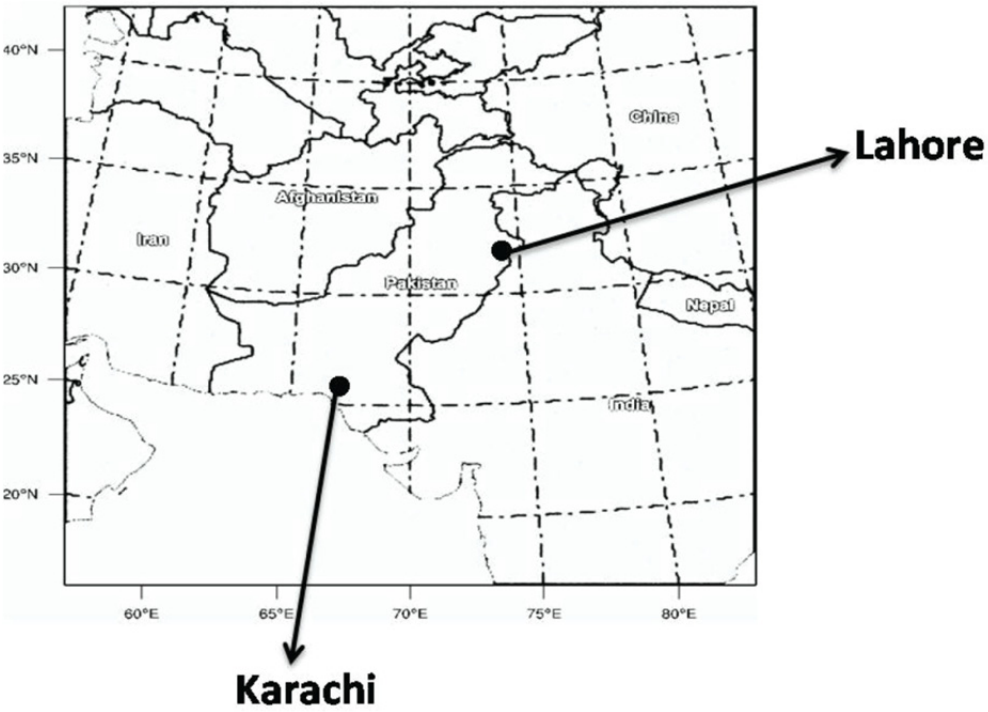

Fig. 1. Model domain and geographical location of Pakistan. The two largest cities of Pakistan, Lahore and Karachi, are indicated.

model has fully coupled meteorology-aerosols-radiation-clouds (Grell *et al.*, 2005; Fast *et al.*, 2006; Gustafson *et al.*, 2007; Chapman *et al.*, 2009). The gas-phase chemistry scheme used in this work is the CBM-Z mechanism (Zaveri and Peters, 1999) with photolysis rates calculated using the Fast-J scheme (Wild *et al.*, 2000). The aerosol module is the Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri *et al.*, 2008) with eight size bins (0.039–0.078, 0.078–0.156, 0.156–0.3125, 0.3125–0.625, 0.625–1.25, 1.25–2.5, 2.5–5.0, 5.0–10 μm). Aerosol particles in each size bin are considered to be internally mixed. Simulated aerosol species in MOSAIC include SO_4^{2-}, NO_3^-, NH_4^+, OC, and BC. The WRF-Chem model considers the interactions of the aerosol radiative effects with meteorological fields. The Goddard shortwave radiative transfer model (Chou *et al.*, 1998) and the Rapid Radiative Transfer Model Longwave Radiation scheme (Mlawer *et al.*, 1997) are used to calculate the aerosol direct radiative effects. The modified Purdue Lin microphysics module by Lin *et al.* (1983) is used for cloud microphysics.

Emissions Inventories

For simulation of aerosol precursors and all aerosol species in the WRF-Chem model, anthropogenic emissions for NO_x, CO, non-methane volatile organic compounds (NMVOCs), SO_2, BC, and OC are taken from the Intercontinental Chemical Transport Experiment-Phase B (INTEX-B) inventory (http://mic.greenresource.cn/intex-b2006) for year 2006. The INTEX-B inventory has a horizontal resolution of 0.5 degree. Emissions of NH_3 are taken from Streets *et al.* (2003). Table 1 summarizes anthropogenic emissions from different sectors (power, industry, residential, and transportation) in Pakistan. Summed over Pakistan (20–40°N, 60–80°E), anthropogenic emissions of NO_x, CO, NMVOCs, SO_2, NH_3, BC, and OC are 2.4 Tg N/yr, 28.8 Tg CO/yr, 5.2 Tg C/yr, 4.8 Tg S/yr, 3.4 Tg N/yr, 0.2 Tg C/yr, and 0.7 Tg C/yr, respectively, in year 2006. Industry and transportation are the two major emission sources of NO_x, accounting for 39.6% and 43.2% of total NO_x emission in Pakistan, respectively. Residential activities are the largest sources of emissions of CO, NMVOCs, BC, and OC, which contribute, respectively, 62.9%, 43.3%, 58.7%, and 96.3% to the annual total emissions of these species. Industry has the largest contribution to SO_2 emissions (accounting for 75.6% of the total). Emissions of NH_3 in Pakistan are mainly from agricultural sector. Fig. 2 shows the horizontal distributions of annual anthropogenic emissions of SO_2, NO_x, NH_3, BC and OC over the simulation domain. Emissions of all species have the same spatial pattern, with highest emissions over northeastern, central, and southeastern Pakistan. Note that because of the lack of information about seasonal variations in emissions, monthly emissions are the same in our simulations for January, April, July, and October of 2006.

Numerical Experiments

The following WRF-Chem simulations are performed in order to examine the seasonal variations in aerosols and to quantify the impacts of transboundary transport on aerosol concentrations in Pakistan:

(1) CTRL: The control simulation of SO_4^{2-}, NO_3^-, NH_4^+, OC, BC, and $PM_{2.5}$ for 4 months (i.e., January, April, July, and October) of year 2006.

(2) TransB: Sensitivity simulation to examine the impacts of transboundary transport on aerosol concentrations in Pakistan in different seasons. This simulation is the same as the CTRL simulation except that anthropogenic emissions over Pakistan are turned off.

For simulation of each of the four months in year 2006, the model is spinned up for 7 days in simulation CTRL or TransB. Initial and lateral boundary conditions for meteorological parameters are taken from the NCEP FNL data with horizontal resolution of 1 degree and temporal resolution of 6 h. (http://rda.ucar.edu/datasets/ds083.2/). Initial conditions and boundary conditions for chemical species are taken from the Model for Ozone and Related Chemical Tracers-version 4 (MOZART-4) (Emmons *et al.*, 2010).

MODEL EVALUATION

Evaluation of Meteorological Parameters

The simulated meteorological fields are evaluated

Table 1. Summary of INTEX-B emissions (Gigagram (Gg)/yr) of SO_2, NO_x, CO, NMVOCs, BC, and OC from different sectors for Pakistan. The number in the parentheses is the fraction of emission from the sector in annual total emission of this species.

Species	Power (Gg)	Industry (Gg)	Residential (Gg)	Transportation (Gg)	Total (Gg)
NO_x	96.3	270.5	20.4	293.6	680.7
	(13.5%)	(39.6%)	(3.0%)	(43.2%)	
CO	0.0	1009.5	4637.9	1730.5	7377.9
	(0%)	(13.7%)	(62.9%)	(23.5%)	
NMVOCs	1.6	232.9	609.9	560.8	1405.3
	(0.1%)	(16.5%)	(43.3%)	(39.9%)	
SO_2	421.3	2179.8	70.1	210.7	2881.9
	(14.6%)	(75.6%)	(2.4%)	(7.3%)	
BC	0.7	36.3	67.4	10.4	114.8
	(0.6%)	(31.6%)	(58.7%)	(8.7%)	
OC	06	2.8	336.8	9.3	349.4
	(1.7%)	(0.8%)	(96.3%)	(2.6%)	

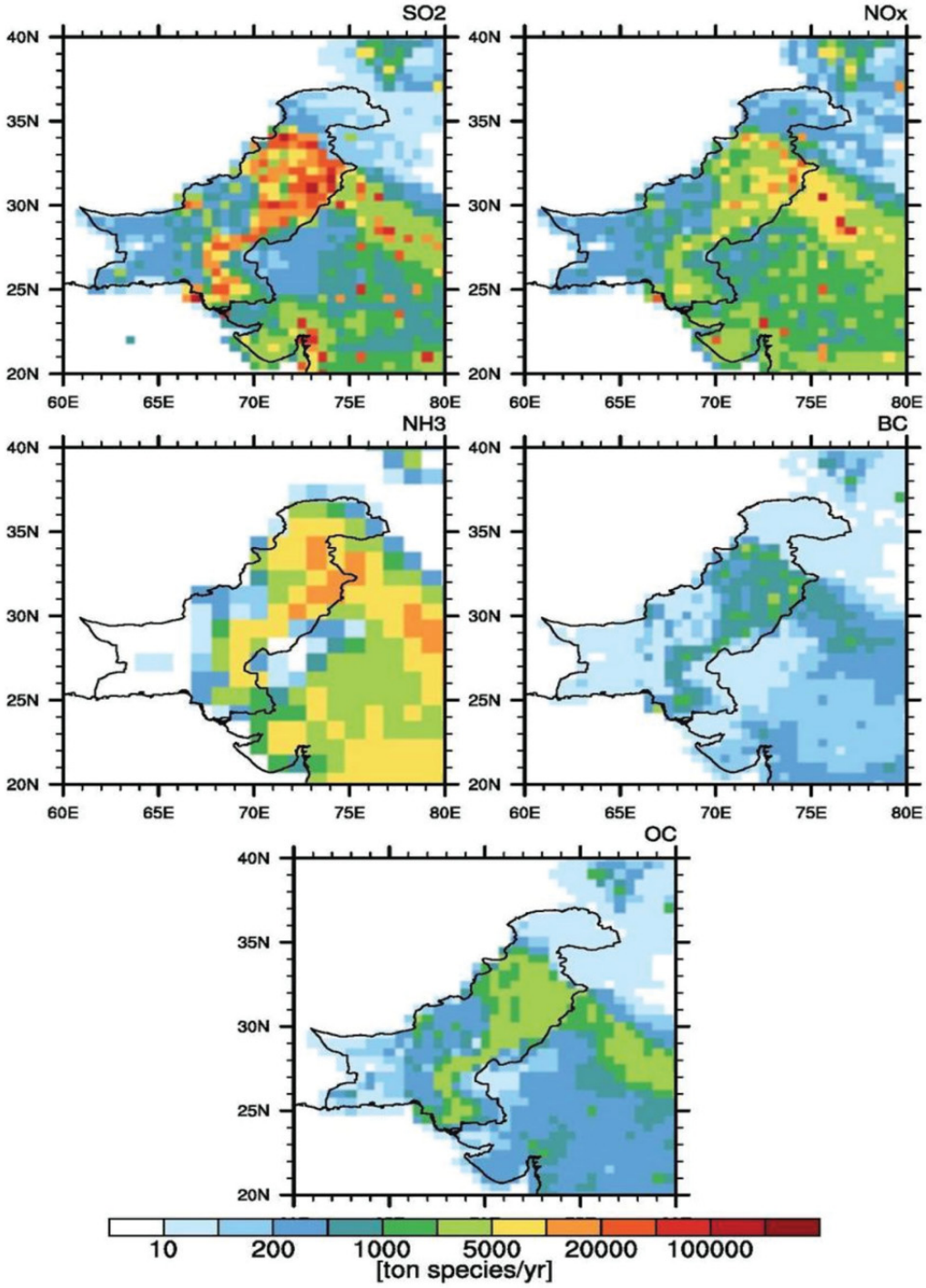

Fig. 2. Spatial distributions of annual mean anthropogenic emissions (tonnes/yr) of SO_2, NO_x, NH_3, BC, and OC over the model domain.

against the reanalysis data. We compare modeled monthly mean surface-air temperature, relative humidity (RH), and wind components with NCEP FNL datasets, which are available at the horizontal resolution of 1 degree and the temporal resolution of 6 h. Simulated precipitation is also compared with datasets from the Tropical Rainfall Measuring Mission (TRMM). We use TRMM Level-3 monthly products available at http://gdata1.sci.gsfc.nasa.gov/daac-bin/G3/gui.cgi?instance_id=TRMM _Monthly. We also quantify the model performance using statistical parameters of mean bias (MB), normalized mean bias (NMB), and correlation coefficient (R) (Table 2). These statistics are calculated using the simulated and reanalyzed meteorological parameters that are averaged over the model domain and are sampled every 6 hours (to be consistent with the temporal resolution of NCEP FNL datasets).

Fig. 3 compares simulated and reanalyzed monthly mean surface-air temperature for January, April, July, and October

of 2006. Surface-air temperatures are higher in central and southern Pakistan than in northern regions in all months, with the maximum temperatures exceeding 35°C in July. The model captures well the magnitudes and distributions of temperatures, with monthly MBs of 0.6°C, 2.2°C, 0.8°C, and –0.01°C and NMBs of 9.4%, 12.4%, 3.2%, and –0.05% in January, April, July, and October, respectively.

Fig. 4 shows simulated and reanalyzed monthly mean surface-layer RH for January, April, July, and October of 2006. The simulated RH is generally high (> 40%) throughout the year in northern and southern parts of Pakistan. The high RH in northern Pakistan is associated with precipitation in that region, and the high RH in southern Pakistan is caused by the low latitudes and moist air from the nearby ocean. RH is well simulated with monthly MBs of 0.8%, –4.6%, –4.1%, and –2.5% and NMBs of 1.6%, –10.5%, –7.7% and –5.4% for January, April, July, and October (Table 2), respectively. The bias is relatively large in April because the absolute value of RH averaged over the model domain is the smallest among the four months.

Fig. 5 shows the simulated wind vectors over the model domain. Winds in northeastern and northern Pakistan are generally smaller than those over the Arabian Sea. In the month of July, strong southerlies prevail in southern and central Pakistan, which are associated with the South Asian summer monsoon. The comparisons of simulated and reanalyzed monthly mean zonal and meridional wind components at surface level are shown in Fig. 6. Against the NCEP datasets for January, April, July and October, the zonal wind is simulated with monthly MBs of –0.01, 0.2, 0.1, and –0.01 m/s while the meridional wind has monthly MBs of –0.2, 0.2, –0.05, and 0.1 m/s (Table 2), respectively. Because of the offsets of positive and negative winds, the absolute values of wind speed are small, leading to large NMBs shown in Table 2. The simulated winds correlate well with reanalyzed winds, with correlation coefficients in the range of 0.84–0.95 (Table 2).

Fig. 7 shows spatial distributions of monthly accumulated

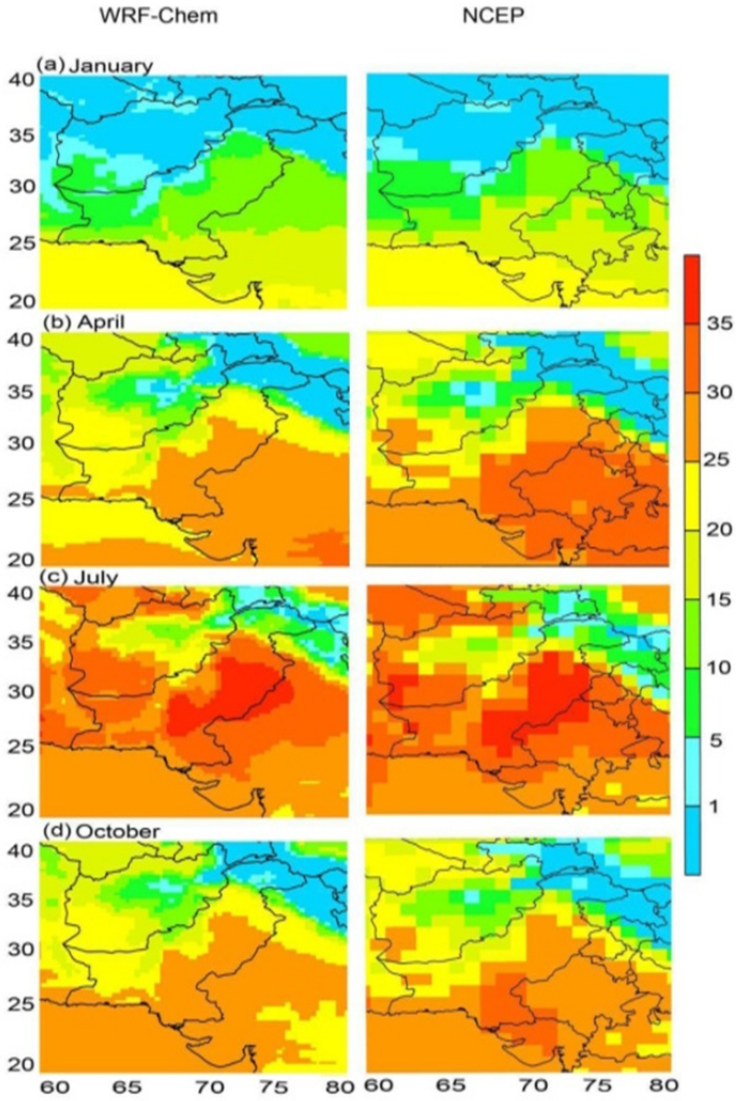

Fig. 3. The monthly mean surface air temperature (°C) from the CTRL simulation (left column) and the NCEP FNL datasets (right column) over the model domain in January, April, July, and October of year 2006.

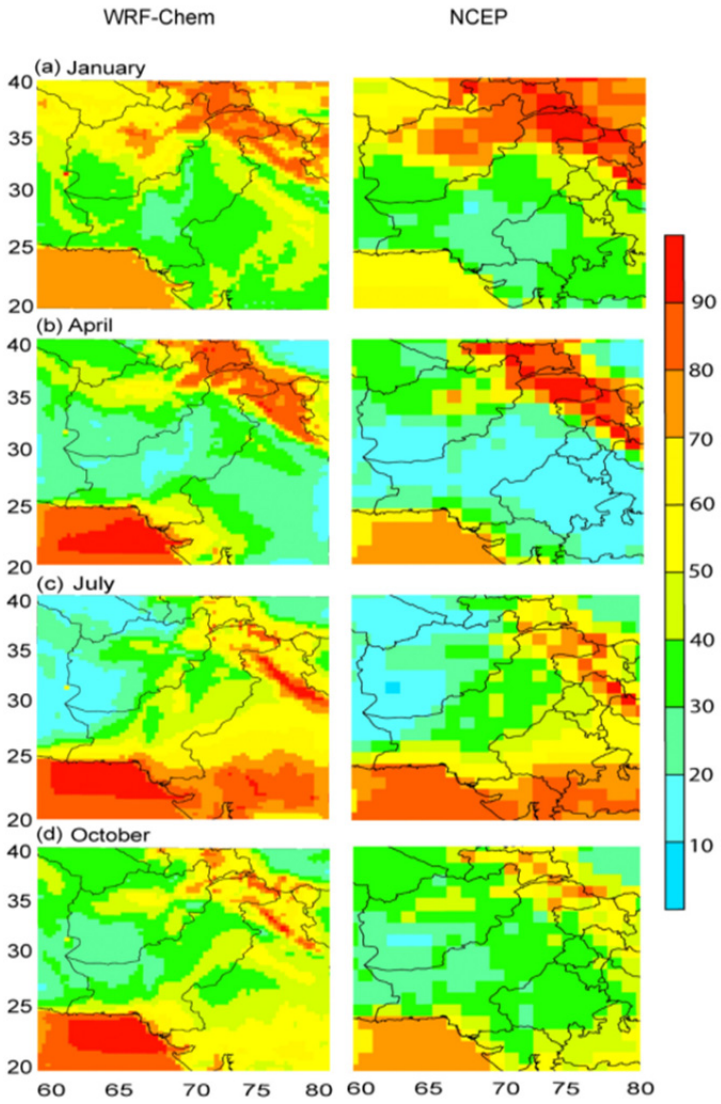

Fig. 4. The monthly mean surface relative humidity (%) from the CTRL simulation (left column) and the NCEP FNL datasets (right column) over the model domain in January, April, July, and October of year 2006.

precipitation from simulation as well as the NCEP and TRMM datasets. The largest precipitation of exceeding 300 mm occurs over the northeastern Pakistan in the month of July. The total rainfall in January also exceeds 200 mm over northern Pakistan because of westerlies over the high mountains. The model captures very well the magnitude and seasonal variation of precipitation over the model domain.

Evaluation of Chemical Parameters

In Pakistan, measurements of aerosols are very scarce for the entire country. Limited ground-based observations are available from the two largest cities in Pakistan: Lahore (Lodhi *et al.*, 2009) and Karachi (Mansha *et al.*, 2012). The geographical locations of these sites are shown in Fig. 1 and Table 3 summarizes the ground-based measurements we collected from the literature.

As shown in Table 3, measured aerosol concentrations vary significantly with season and year. Observed concentrations at Lahore and Karachi, exhibited maximum in winter and minimum in summer (Harrison *et al.*, 1997; Hameed *et al.*, 2000; Rattigan *et al.*, 2002; Biswas *et al.*, 2008; Lodhi *et al.*, 2009; Economic Survey 2009–2010; Alam *et al.*, 2011a; Stone *et al.*, 2012; Mansha *et al.*, 2012).

We evaluate first the simulated aerosol concentrations at Lahore. Due to the unavailability of measurements for April, July, and October of 2006, the model results are compared with observations at Lahore for January 2006 (Fig. 8), with measurements taken from Lodhi *et al.* (2009). Observed SO_4^{2-} concentrations from years 1999 and 2000 and observed NO_3^- from year 1999, which were reported in Rattigan *et al.* (2002) and Hameed *et al.* (2000), are also shown in Fig. 8 to demonstrate the magnitudes of observed concentrations. The comparison shows that simulated SO_4^{2-} concentrations agree closely with observed values for January of 2006. The model generally underestimates concentrations of NO_3^-, NH_4^+, OC, and $PM_{2.5}$ in January of 2006. Averaged over hours with measurements available, concentrations of NO_3^-, NH_4^+, OC, and $PM_{2.5}$ are simulated to be 14.0, 5.5,

Table 2. Mean bias (MB) and normalized mean bias (NMB) of the simulated surface-layer meteorological parameters against the NCEP reanalyzed datasets for January, April, July, and October of 2006. The correlation coefficient (*R*) between the simulated and reanalyzed datasets is also shown. These statistics are calculated using the simulated and reanalyzed meteorological parameters that are averaged over the model domain and are sampled every 6 hours (to be consistent with the temporal resolution of NCEP FNL datasets).

Variable		Mean Bias (MB)	Normalized Mean Bias (NMB, %)	*R*
Temperature	January	0.6°C	9.4%	0.98
	April	2.2°C	12.4%	0.99
	July	0.8°C	3.2%	0.99
	October	–0.01°C	–0.05%	0.98
Relative Humidity	January	0.8%	1.6%	0.98
	April	–4.6%	–10.5%	0.98
	July	–4.1%	–7.7%	0.99
	October	–2.5%	–5.4%	0.99
U–wind	January	–0.01 m/s	–26.0%	0.94
	April	0.2 m/s	12.6%	0.95
	July	0.1 m/s	8.2%	0.94
	October	–0.01 m/s	–1.4%	0.91
V-wind	January	–0.2 m/s	58.5%	0.94
	April	0.2 m/s	–42.9%	0.89
	July	–0.05 m/s	–19.6%	0.84
	October	0.1 m/s	–35.3%	0.87

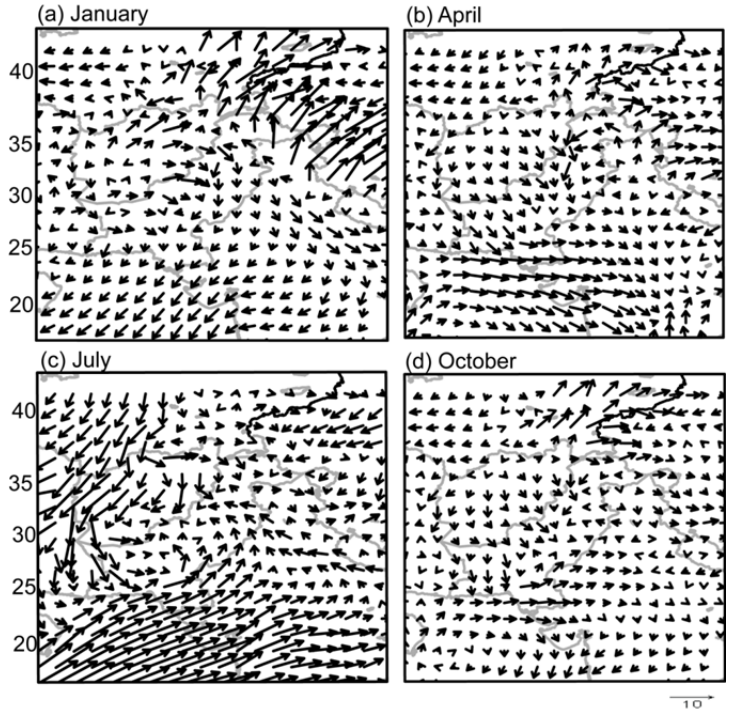

Fig. 5. Simulated monthly mean wind vectors over the model domain during (a) January, (b) April, (c) July, and (d) October of year 2006. Wind speed and wind direction are represented by arrows. The length of arrow of wind speed of 10 m/s is indicated below the bottom right panel.

15.4, and 48.0 μg/m^3, while observed concentrations are 25.2, 12.6, 57.1, and 142.2 μg/m^3, respectively. These model biases suggest that there might be some uncertainties in anthropogenic emissions.

We then evaluate simulated aerosol concentrations at Karachi. Measured concentrations of SO_4^{2-}, NO_3^-, and $PM_{2.5}$ at Karachi during January, April, July, and October of 2006 are taken from Mansha *et al.* (2012). Fig. 9 compares simulated and observed concentrations for dates with measurements available. Simulated SO_4^{2-} concentrations peak in July, but the observed SO_4^{2-} concentrations show maximum in October. Simulated NO_3^- concentrations agree with measurements in January and April, and the model results show large low biases in July and October.

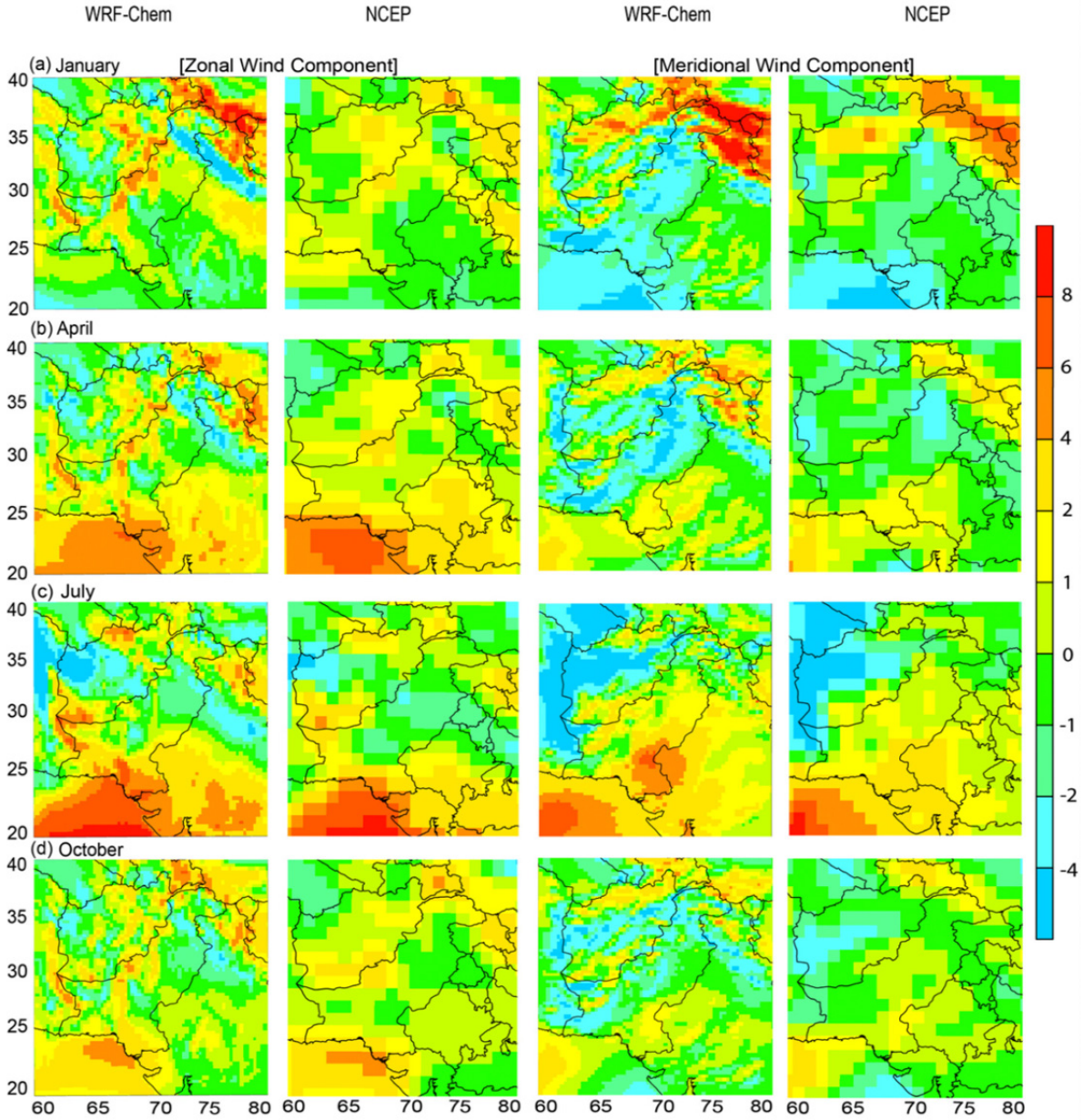

Fig. 6. The monthly mean surface zonal and meridional wind components (m/s) from the CTRL simulation (1st & 3rd columns) and the NCEP FNL datasets (2nd & 4th columns) over the model domain in January, April, July, and October of year 2006.

Simulated concentrations of NO_3^- over Karachi during the months of July and October are very low, which can be attributed to several reasons. First, emissions of all chemical species over Karachi are lower than emissions over northeastern Pakistan (Fig. 2). Second, strong southerlies from Arabian Sea in July (Fig. 5) transport NO_3^- from Karachi to downwind areas. Third, high temperatures during July and October in southern Pakistan are not favorable for NO_3^- formation. While the simulated NO_3^- concentrations are practically zero in July and October, measured nitrate concentrations are in the range of 5–50 μg/m^3 in these two months. With respect to $PM_{2.5}$, simulated concentrations are about 20 μg/m^3 in all seasons. It should be noted that observed $PM_{2.5}$ concentrations are the highest in January (100–120 μg/m^3), followed by April (50–80 μg/m^3), July (40–50 μg/m^3), and October (about 40 μg/m^3).

Evaluation of Aerosol Optical Depth (AOD)

Satellite measurements provide large spatial and temporal coverage and can be used to evaluate simulated aerosols by examining aerosol optical depth. We use retrieved AOD from the Moderate-resolution Imaging Spectroradiometer (MODIS) Terra satellite at the wavelength of 550 nm for months of January, April, July, and October in 2006. The datasets are level 3 monthly products downloaded from NASA Giovanni website (http://disc.sci.gsfc.nasa.gov/giovanni). We also use retrieved AOD from the Aerosol Robotic NETwork (AERONET) (Holben *et al.*, 1998). Currently

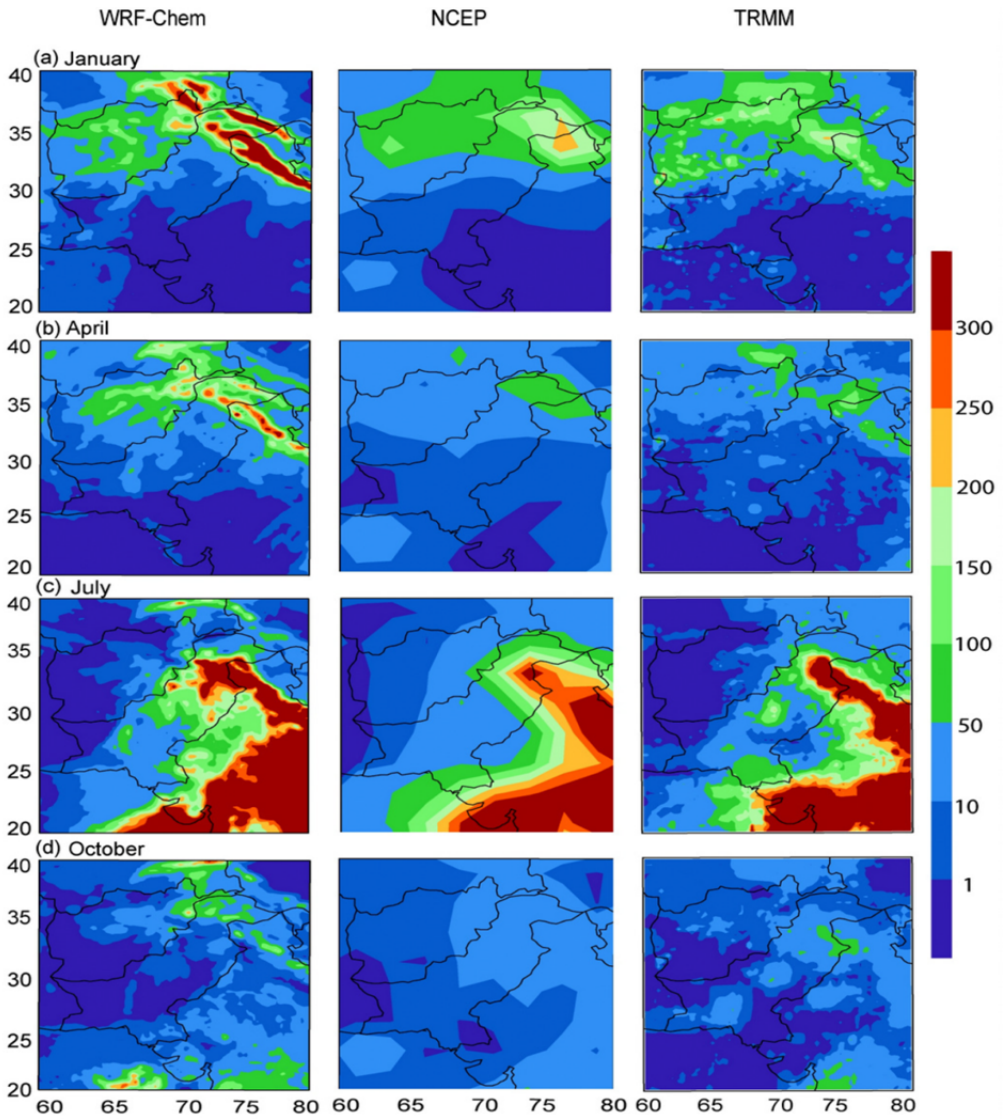

Fig. 7. The monthly accumulated precipitation (mm) simulated from the CTRL simulation (left column), the NCEP FNL datasets (middle column), and the TRMM datasets (right column) over the model domain in January, April, July, and October of year 2006.

there are 2 AERONET sites in Pakistan and their levels 1.0, 1.5 and 2.0 products of AOD are available. Retrieved level 2.0 AOD values at Lahore during January 2007–October 2012 and at Karachi, October 2006–October 2012 are used in model evaluation. The optical properties of aerosols simulated in the WRF-Chem model are at the wavelengths of 300, 400, 600, and 999 nm. Since AOD from AERONET and satellite are at the wavelengths of 500 nm and 550 nm, respectively, simulated AOD values are interpolated to 500 nm and 550 nm following the Angström power law (http://disc.sci.gsfc.nasa.gov/data-holdings/PIP/aerosol_angstrom _exponent.shtml).

Fig. 10 shows the simulated monthly mean AOD in the CTRL simulation and MODIS retrieved AOD for 2006. Simulated AOD values are the averages over hours 10 and 11 AM, since the Terra local equatorial crossing time is approximately 10:30 AM (http://nsidc.org/data/modis/terra_aqua_differences/). Simulated monthly mean AOD values are in the range of 0.3–0.5 in January, and are 0.5–0.7 in northern Pakistan and 0.7–0.9 in southern Pakistan in April. Simulated AOD exceeds 0.9 over eastern Pakistan in July as a result of the high RH of 40–60% in this month (Fig. 4). In October, simulated monthly mean AOD values are 0.5–0.7 over eastern Pakistan. The simulated seasonal variations and magnitudes of AOD agree closely with the MODIS measurements. Both simulated and observed AOD values peak in July although simulated $PM_{2.5}$ concentrations are the lowest in this month (Fig. 12). However, the model slightly overestimates AOD in April as compared to the MODIS measurements. Note that mineral dust aerosol contributes largely to AOD in the months of April and July. Kaskaoutis *et al.* (2011) reported that dust load is high over northwestern India and Pakistan in April to July due to the long range transport of dust from the Arabian Peninsula and the Middle East. Dey *et al.* (2004) also reported that high AOD values were observed over Indian

Table 3. Ground-based measurements of aerosols in Pakistan reported in the literature.

Location	Period	Concentration (μg/m^3)	Reference
Observed concentrations of SO_4^{2-} aerosol			
Lahore	October, 1992–October, 1993	18.4	Harrison *et al*. (1997)
(74°22′E, 31°32′N)	December, 1998–January, 1999	17–99	Hameed *et al*. (2000)
	December, 1999–January, 2000	4–141	Rattigan *et al*. (2002)
	June, 2007	0.5–7.2	Stone *et al*. (2012)
	December, 2007	0.5–10.6	Stone *et al*. (2012)
	November, 2005–March, 2006	3.3–59	Lodhi *et al*. (2009)
	December, 2005–February, 2006	19.2	Lodhi *et al*. (2009)
Karachi	January–March, 2006–2008	0.9–7.5	Mansha *et al*. (2012)
(67°E, 24°53′N)	April–June, 2006–2008	1.0–7.6	Mansha *et al*. (2012)
	July–September, 2006–2008	1.8–8.8	Mansha *et al*. (2012)
	October–December, 2006–2008	3.6–10.5	Mansha *et al*. (2012)
	2005–2010	53.2	Economic Survey (2009–2010)
Observed concentrations of NO_3^- aerosol			
Lahore	October, 1992–October, 1993	12.8	Harrison *et al*. (1997)
(74°22′E, 31°32′N)	December, 1998–January, 1999	22.8–56.5	Hameed *et al*. (2000)
	December, 1999–January, 2000	3–74.5	Rattigan *et al*. (2002)
	June, 2007	0.3–2.9	Stone *et al*. (2012)
	December, 2007	0.8–16.0	Stone *et al*. (2012)
	November, 2005–March, 2006	2.10–73	Lodhi *et al*. (2009)
Karachi	January–March, 2006–2008	0.6–6.28	Mansha *et al*. (2012)
(67°E, 24°53′N)	April–June, 2006–2008	1.8–4.7	Mansha *et al*. (2012)
	July–September, 2006–2008	12.4–14.3	Mansha *et al*. (2012)
	October–December, 2006–2008	13.1–15.4	Mansha *et al*. (2012)
Observed concentrations of BC aerosol			
Lahore	November, 2005–January, 2006	21.7	Hussain *et al*. (2007)
(74°22′E, 31°32′N)	November, 2005	17.3	Dutkiewicz *et al*. (2009)
	January, 2006	21.7	Dutkiewicz *et al*. (2009)
Karachi	November 2006–February 2007	10	Dutkiewicz *et al*. (2009)
(67°E, 24°53′N)	June–September, 2007	2	Dutkiewicz *et al*. (2009)
Pakistan	April, 2002–2005	10	Hopke *et al.* (2008)
Observed $PM_{2.5}$ concentrations			
Lahore	November, 2005–March, 2006	53–476	Lodhi *et al*. (2009)
(74°22′E, 31°32′N)	December, 2005–February, 2006	209	Biswas *et al*. (2008)
	June, 2007	15.1–106	Stone *et al*. (2012)
	December, 2007	28.1–280.6	Stone *et al*. (2012)
	March–April, 2010	91	Alam *et al*. (2011a)
	2005–2010	121.8	Economic Survey (2009–2010)
Peshawar	March–April, 2010	160	Alam *et al*. (2011a)
(71°37′E, 34°02′N)	2005–2010	70.2	Economic Survey (2009–2010)
Islamabad	March–April, 2010	140	Alam *et al*. (2011a)
(73°10′E,33°40′N)	2005–2010	73.0	Economic Survey (2009–2010)
Karachi	January–March, 2006–2008	98.44	Mansha *et al*. (2012)
(67°E, 24°53′N)	April–June, 2006–08	55.89	Mansha *et al*. (2012)
	July–September, 2006–2008	46	Mansha *et al*. (2012)
	October–December, 2006–2008	116.97	Mansha *et al*. (2012)
	2005–2010	53.2	Economic Survey (2009–2010)
Quetta	2005–2010	47.1	Economic Survey (2009–2010)
(66°55′E, 30°15′N)			
Pakistan	April, 2002–2005	35	Hopke *et al*. (2008)

subcontinent during June to August, as a result of the strong southwesterlies that transported dust from the Thar and the Arabian deserts.

Fig. 11 compares simulated and AERONET AOD values at 500 nm for Lahore and Karachi. The AERONET datasets are only available for October 2006 for Karachi; therefore we also compare the simulated AOD values with measurements from years 2007–2012. Modeled AOD values over Lahore

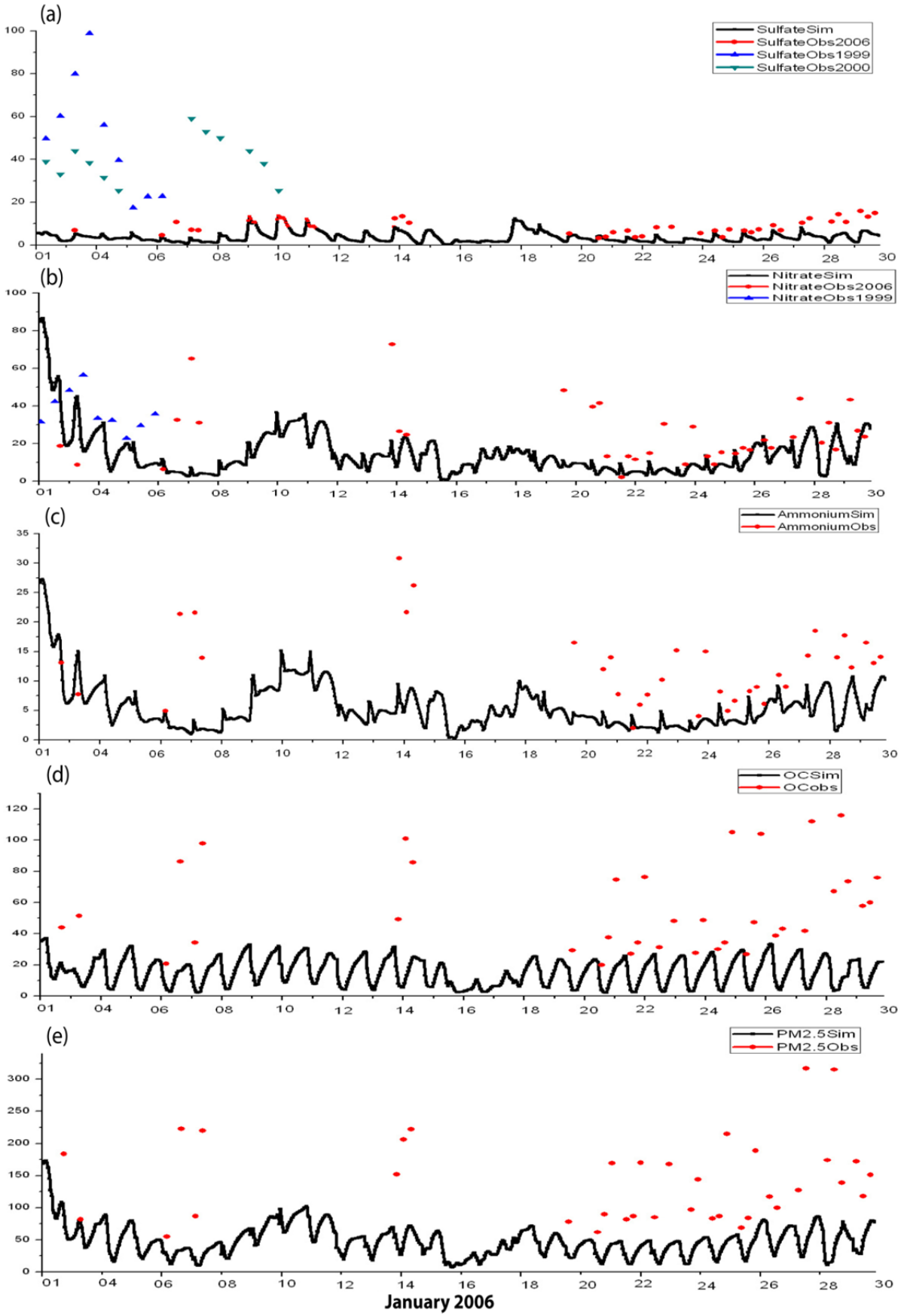

Fig. 8. Simulated and observed daily variations in surface-layer concentrations (μg/m^3) of (a) SO_4^{2-}, (b) NO_3^-, (c) NH_4^+, (d) OC, and (e) $PM_{2.5}$ at Lahore in January of 2006.

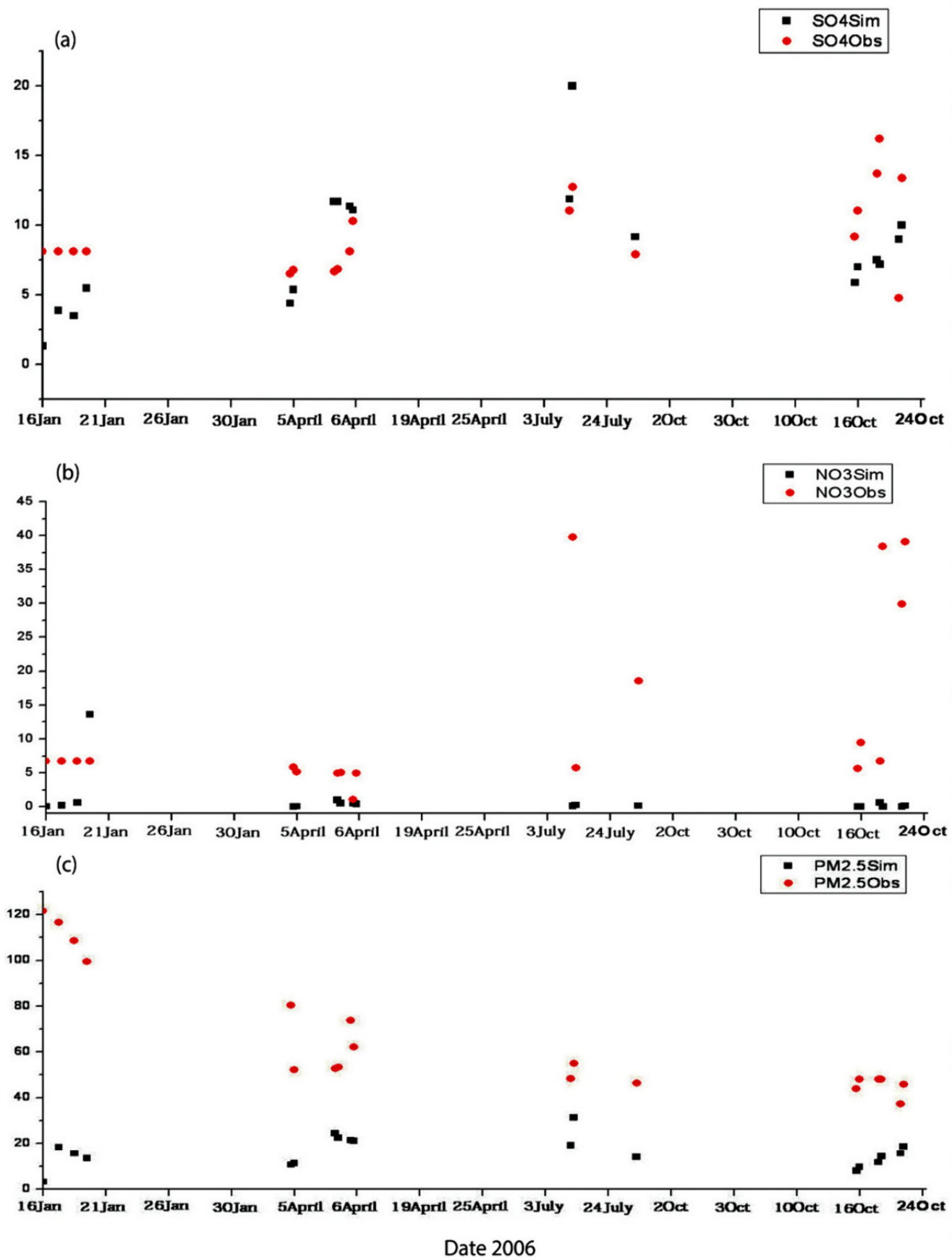

Fig. 9. Simulated and observed surface-layer concentrations (μg/m^3) of (a) SO_4^{2-}, (b) NO_3^-, and (c) $PM_{2.5}$ at Karachi in January, April, July, and October of 2006.

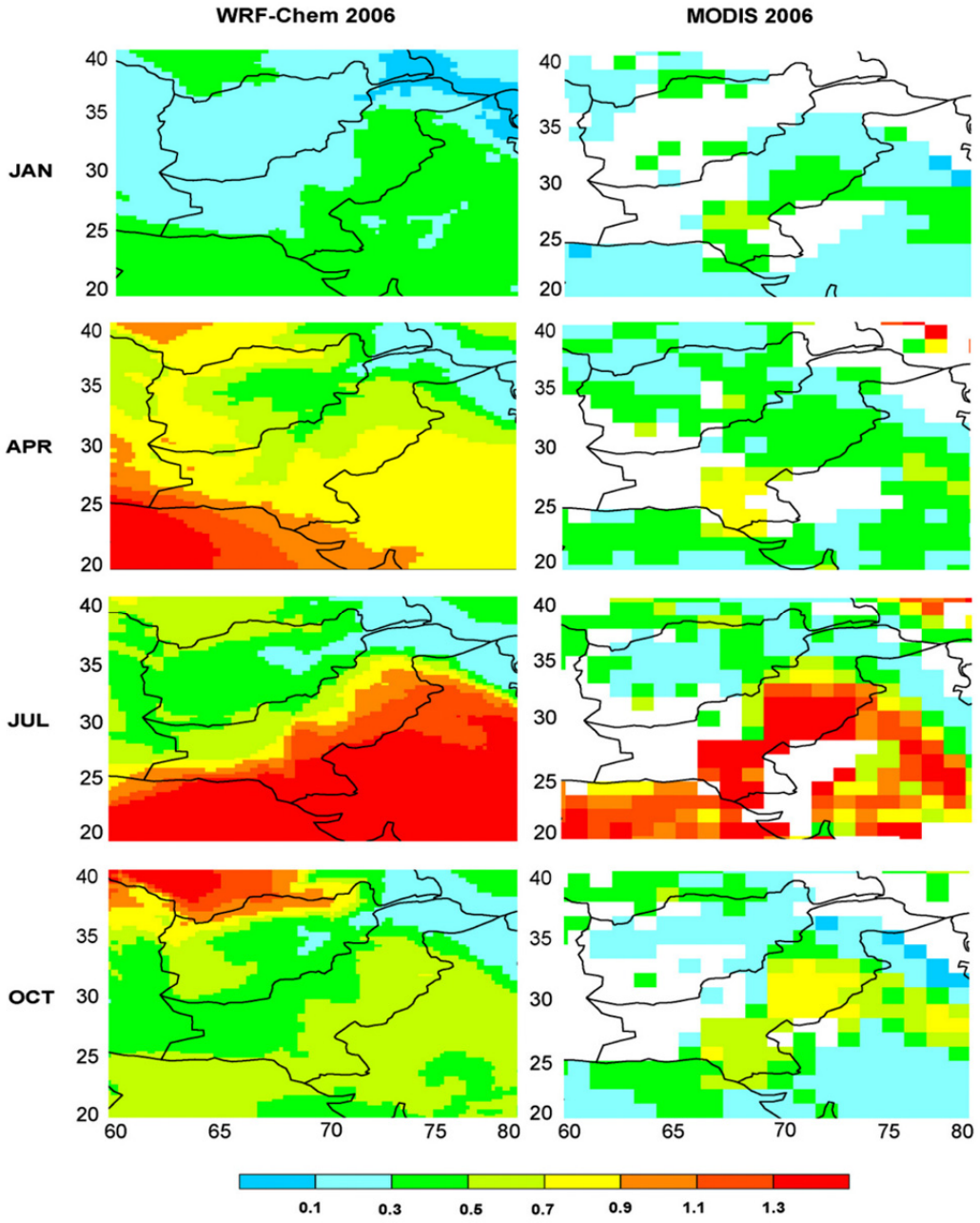

Fig. 10. Comparisons of simulated monthly mean AOD (left column) with the MODIS AOD (right column) for year 2006. Regions with white color indicate places with no MODIS AOD datasets available.

are within the range of AERONET retrievals in January, April, July, and October. In Karachi, simulated AOD values agree with AERONET observations in January and October, but the model overestimates AOD values by about 0.4–0.5 in April and July. Alam *et al.* (2010b) reported that AOD values were high in the southern coastal areas of Pakistan during the humid summer season due to hygroscopic aerosols as well as the presence of mineral dust.

SEASONAL VARIATION OF AEROSOLS IN PAKISTAN

The simulated horizontal distributions of monthly mean concentrations of aerosols are shown in Fig. 12 for January, April, July, and October to represent aerosol concentrations in four seasons in Pakistan. Over eastern Pakistan, SO_4^{2-} aerosol exhibits maximum concentrations of 5–10 μg/m^3 in July and October and of 3–5 μg/m^3 in January and April. Strong photochemistry facilitates maximum SO_4^{2-} formation in July almost all over Pakistan. High sulfate concentrations in summer are attributed to more rapid oxidation of SO_2 to sulfate in warmer months because of higher rate constants and higher oxidant concentrations (Aw and Kleeman, 2003; Dawson *et al.*, 2007; Kleeman, 2007). Simulated concentrations of NO_3^- are generally higher than those of SO_4^{2-} in northeastern Pakistan. The highest NO_3^-

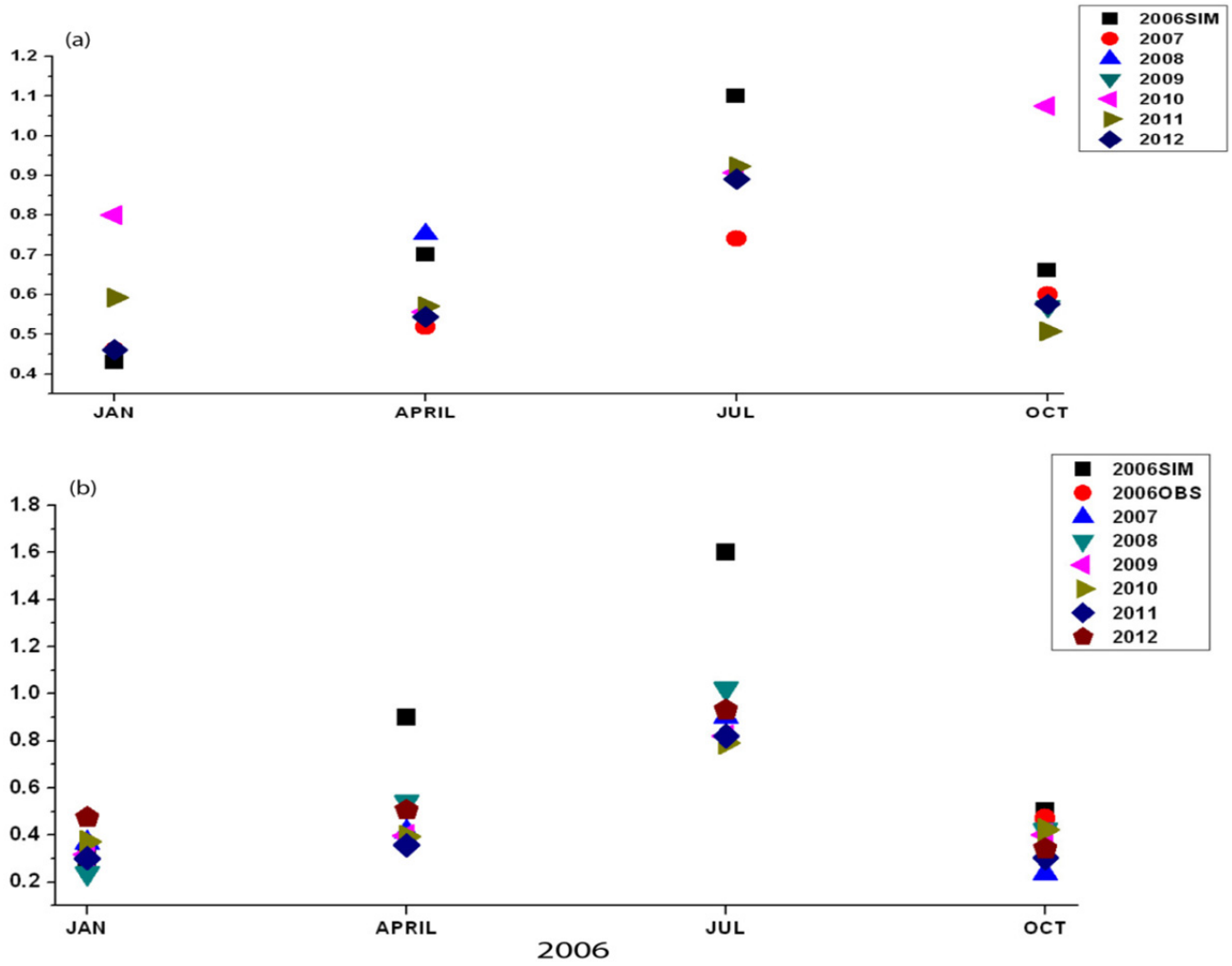

Fig. 11. Comparison of simulated AOD and AERONET retrieved AOD at 500 nm for (a) Lahore and (b) Karachi.

concentrations of 30–50 μg/m^3 are simulated in April and October, as a result of the relatively low temperatures and precipitation. In contrast, high temperatures and large rainfall lead to maximum NO_3^- concentrations of about 25–30 μg/m^3 in July. The simulated high nitrate concentrations are likely caused by the overestimate of NO_x emissions. Ghude *et al.* (2013) reported the overestimation of NO_x emissions in the INTEX-B inventory for the western and eastern Indo-Gangetic region using the WRF-Chem model with an iterative inverse technique. Simulated highest BC (or OC) concentrations in northeastern Pakistan are 5–10 μg/m^3 (or 10–20 μg/m^3) in January and October. Simulated $PM_{2.5}$ concentrations averaged over northeastern Pakistan (71–74.5°E, 28–34°N) are 55, 48.5, 31.5, and 98 μg/m^3 in January, April, July, and October, respectively.

In general, simulated aerosol concentrations are high over eastern Pakistan and aerosol concentrations show strong seasonal variations. Compared to the concentrations in January, aerosol levels in April show reductions but high concentration levels remain over the northeastern Pakistan. In July, the summer monsoon cleans out northeastern part of the Pakistan but pollutants start to build up over the central and southern Pakistan where precipitation is not significant (Fig. 7). During the post monsoon period (October), aerosol concentrations become significant again, with concentrations in October higher than those in January.

Fig. 13 shows the percentages of SO_4^{2-}, NO_3^-, NH_4^+, BC and OC in the total $PM_{2.5}$ concentration over Pakistan for the four months. In January and April, NO_3^- and OC are simulated to have the largest contributions to $PM_{2.5}$ over northeastern and central Pakistan, with NO_3^- accounting for about 35–60% of $PM_{2.5}$ in eastern Pakistan and OC contributing about 25–45% to $PM_{2.5}$ in central Pakistan. In July, SO_4^{2-} is simulated to be the most dominant chemical component of $PM_{2.5}$ all over Pakistan, which contributes to $PM_{2.5}$ by about 30–40% in northern Pakistan and by about 50–90% in southern Pakistan. In October, NO_3^- is simulated to have the largest contribution of 40–50% to $PM_{2.5}$ over eastern Pakistan, and SO_4^{2-} is still the most dominant aerosol species in southern Pakistan.

The large contributions of NO_3^-, OC, and SO_4^{2-} to $PM_{2.5}$ can also be seen in Fig. 14, in which we show the percentages of each anthropogenic aerosol species in the total $PM_{2.5}$ concentration in the two largest cities of Lahore and Karachi. Over Lahore, the most dominant aerosol species are OC (27–32%) and NO_3^- (24–28%) in January and April, SO_4^{2-}

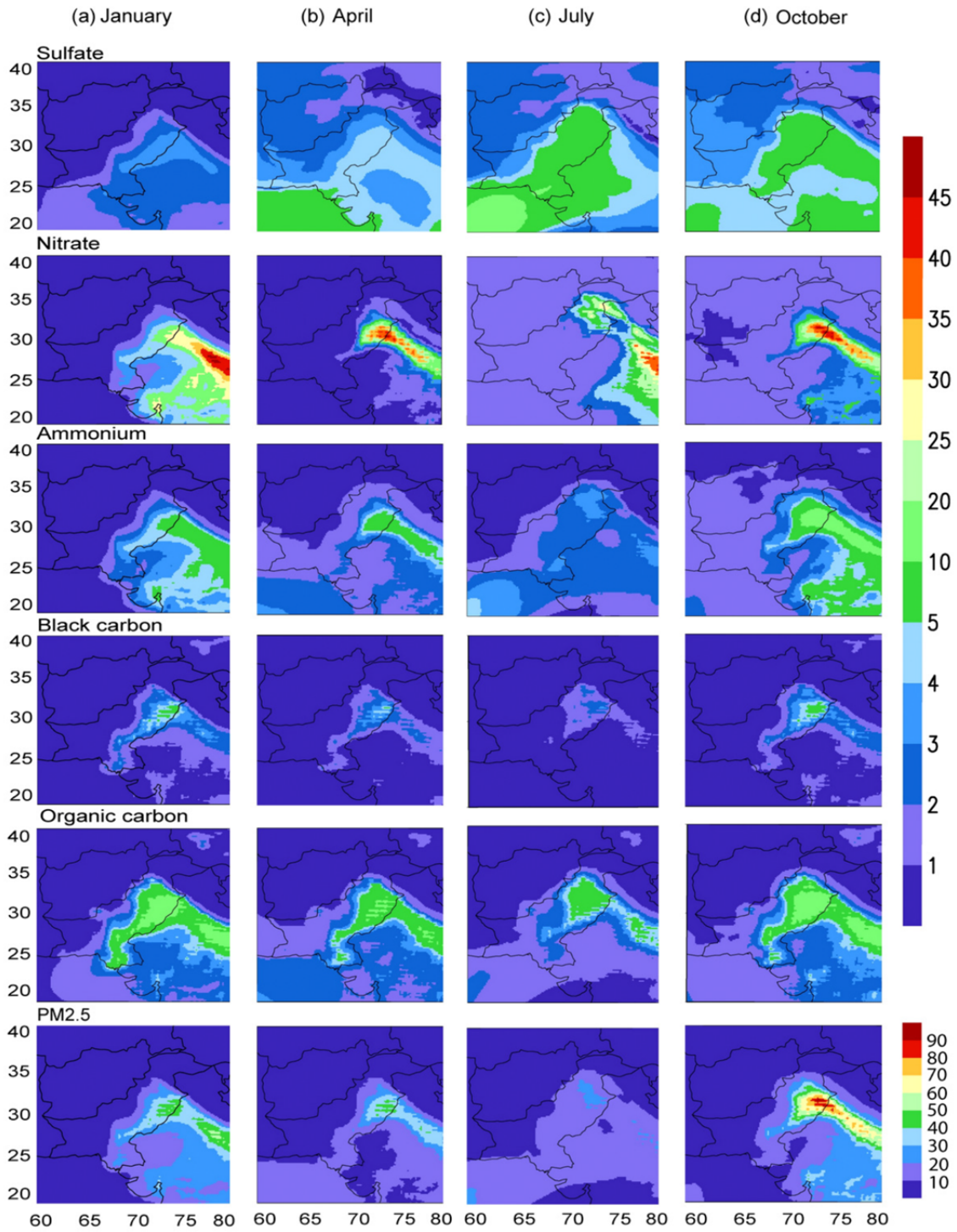

Fig. 12. Simulated monthly mean surface-layer concentrations (μg/m^3) of SO_4^{2-}, NO_3^-, NH_4^+, BC, OC, and $PM_{2.5}$ in the CTRL simulation for (a) January, (b) April, (c) July, and (d) October of 2006.

(31%) and OC (30%) in July, and NO_3^- (38%) in October. Over Karachi, SO_4^{2-} has a dominant contribution to $PM_{2.5}$ by 47%, 66%, and 58% in April, July, and October, respectively.

IMPACTS OF TRANSBOUNDARY TRANSPORT ON AEROSOL LEVELS OVER PAKISTAN

As described in the section of "Numerical Experiments", we performed a simulation experiment TransB (with anthropogenic emissions from Pakistan turned off) in order to examine the impacts of transboundary transport on aerosol concentrations in Pakistan in different seasons. Simulated monthly mean surface-layer concentrations of SO_4^{2-}, NO_3^-, NH_4^+, BC, OC, and $PM_{2.5}$ from TransB are shown in Fig. 15. The contributions of transboundary transport to aerosol levels in Pakistan can also be quantified

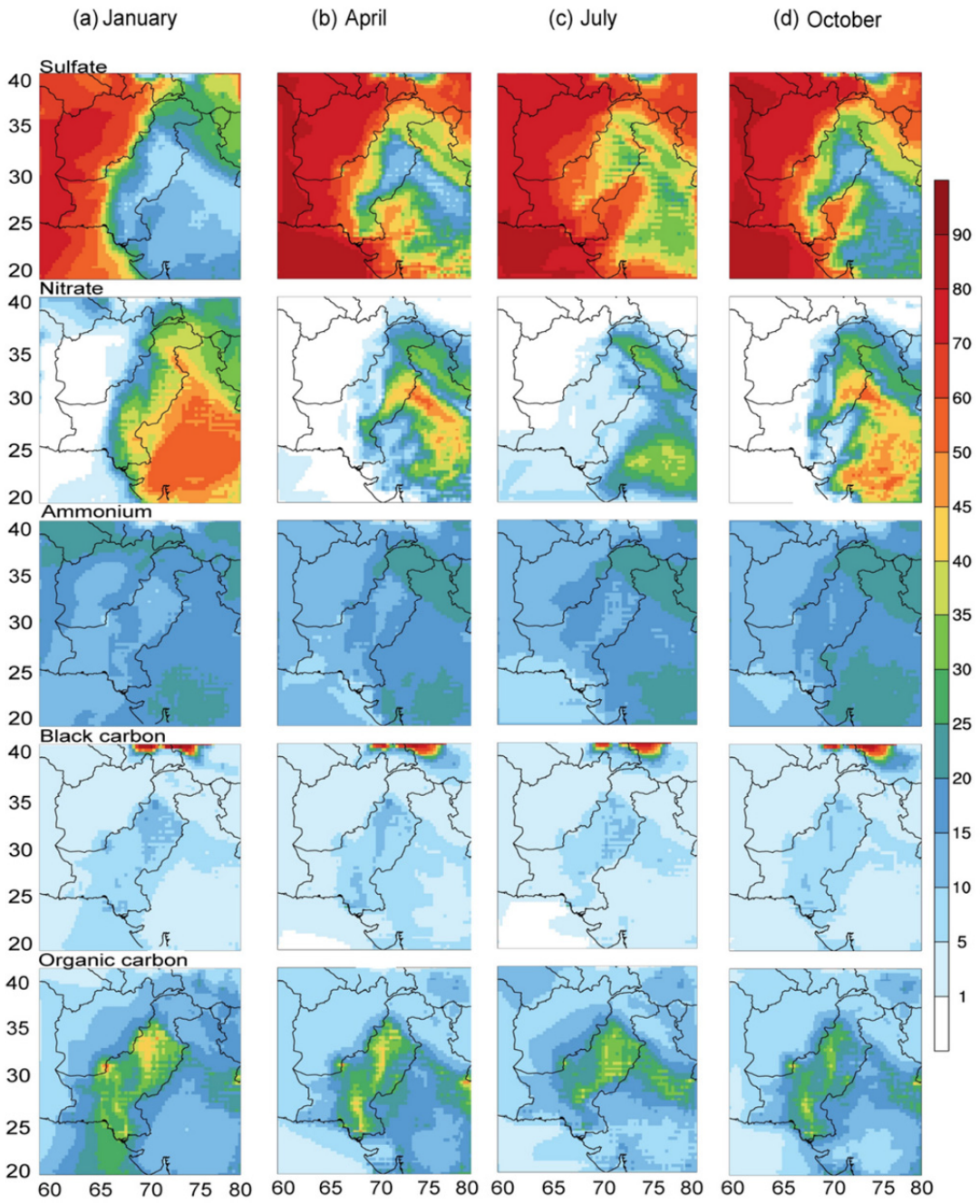

Fig. 13. Simulated percentages (%) of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC in the total $PM_{2.5}$ concentration for (a) January, (b) April, (c) July, and (d) October of year 2006.

by the ratios of aerosol concentrations in TransB to those simulated in the CTRL simulation (Fig. 16). Simulated aerosol concentrations in TransB (Fig. 15) show that the transboundary pollution plays important role in aerosol levels over northeastern Pakistan in all months. In January, easterlies in northeastern Pakistan (Fig. 6) lead to high concentrations of NO_3^-, NH_4^+, and OC transported from India (Fig. 15). As a result, 5–10 μg/m^3 (or 10–20% as shown in Fig. 16) of $PM_{2.5}$ concentration in northeastern Pakistan in January can be attibuted to transboundary transport. During April, transboundary transport contributes 5–10 μg/m^3 (or 10–20%) of $PM_{2.5}$ over the border of northeastern Pakistan. During the month of July, strong southeasterlies associated with the summer monsoon favor the transport of aerosols from India to northeastern Pakistan, but aerosols are washed out by heavy precipitation. Transboundary transport contributes 10–40% of $PM_{2.5}$ over the whole northern Pakistan in July, although the overall $PM_{2.5}$ levels in Pakistan are the lowest in this month (Fig. 12). In October, with prevailing easterlies and low precipitation in northeastern Pakistan and India, 10–35 μg/m^3 (or 10–40%) of $PM_{2.5}$ in northeastern Pakistan are transported from India (Figs. 15 and 16).

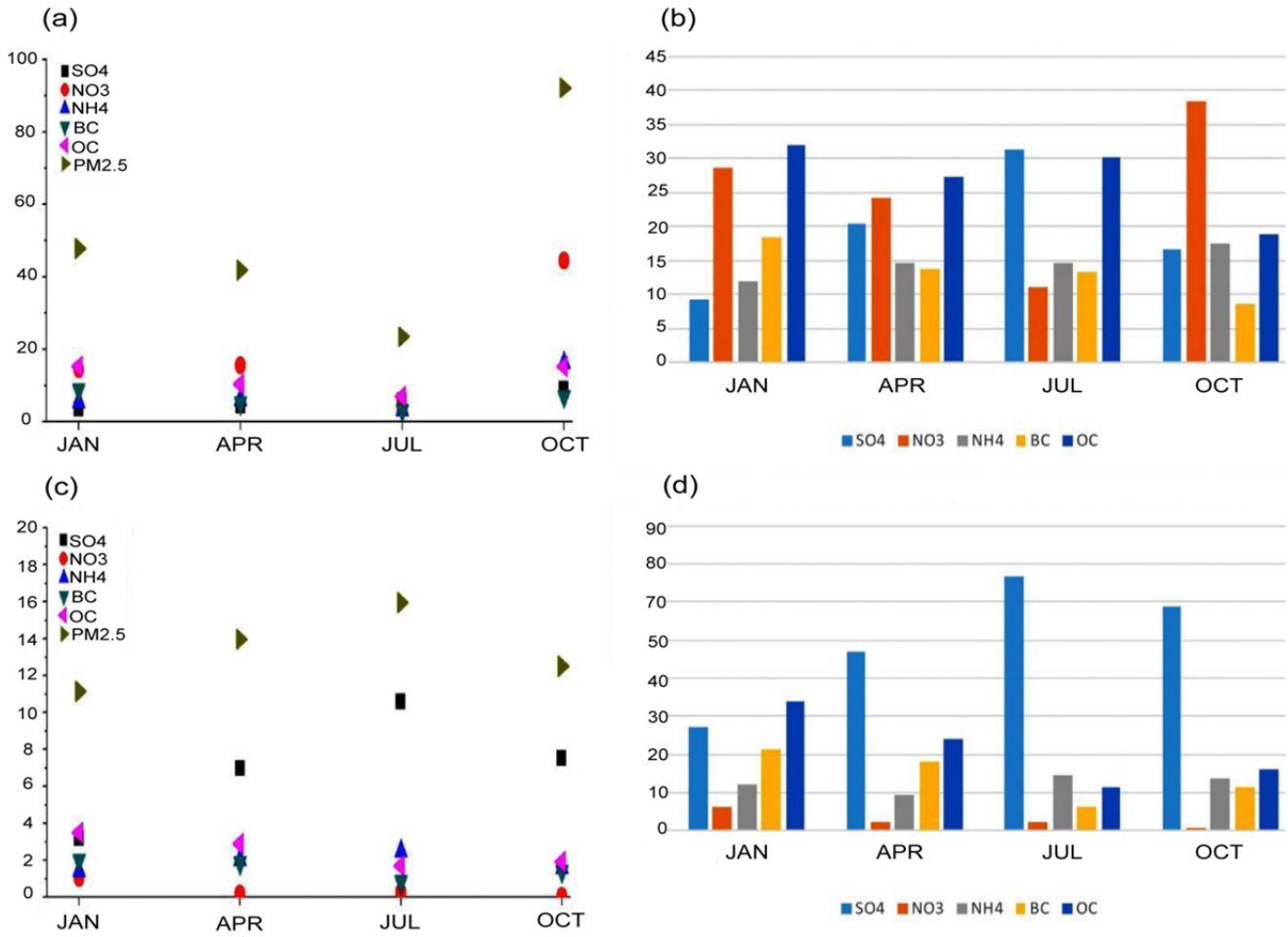

Fig. 14. Left panels: Simulated seasonal variations of concentrations (μg/m^3) of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC at (a) Lahore and (c) Karachi. Right panels: Simulated percentages (%) of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC in the total $PM_{2.5}$ concentration for (b) Lahore and (d) Karachi. These model results are from the CTRL simulation for year 2006.

CONCLUSIONS

We present here a first regional modeling study on the spatial distribution and temporal variation of aerosol concentrations over Pakistan using the WRF-Chem Model. Concentrations of sulfate, nitrate, ammonium, black carbon, organic carbon, and $PM_{2.5}$ are simulated for year 2006 by using anthropogenic emissions from the INTEX-B emissions inventory. Because of the lack of information about seasonal variations in emissions, annual mean emissions are used in our simulations. We examine seasonal variations of aerosols and the impacts of transboundary transport on aerosol levels in Pakistan.

Concentrations of aerosols are simulated for January, April, July, and October of 2006 to represent aerosol concentrations in four seasons in Pakistan. Model simulation shows that concentrations of nitrate, ammonium, black carbon, organic carbon, and $PM_{2.5}$ are the highest in October, followed by those in January, April, and July. The highest concentrations of these aerosol species in October result from the relatively low temperatures that favor nitrate formation as well as the lowest precipitation that leads to the smallest wet deposition of aerosols. The simulated lowest concentrations of nitrate, ammonium, black carbon, organic carbon, and $PM_{2.5}$ in July are attributed to the largest precipitation associated with the South Asian summer monsoon. Note that simulated sulfate aerosol shows different seasonal variations from other aerosol species; simulated sulfate concentrations are the highest in July. In year 2006, simulated $PM_{2.5}$ concentrations averaged over northeastern Pakistan (71–74.5°E, 28–34°N) are 55, 48.5, 31.5, and 98 μg/m^3 in January, April, July, and October, respectively. Over Lahore, the most dominant aerosol species are simulated to be OC (account for 27–32% of $PM_{2.5}$) and NO_3^- (24–28%) in January and April, SO_4^{2-} (31%) and OC (30%) in July, and NO_3^- (38%) in October. Over Karachi, SO_4^{2-} has a dominant contribution to $PM_{2.5}$ by 47%, 66%, and 58% in April, July, and October, respectively.

We also perform a sensitivity simulation with anthropogenic emissions in Pakistan turned off to examine the impacts of transboundary transport on aerosol concentrations in Pakistan in different seasons. Transboundary pollution is simulated to be important over northeastern Pakistan in all months; transboundary transport contributes to $PM_{2.5}$ aerosol levels in northeastern Pakistan by 10–20% in January and April and by 10–40% in July and October. Wind over

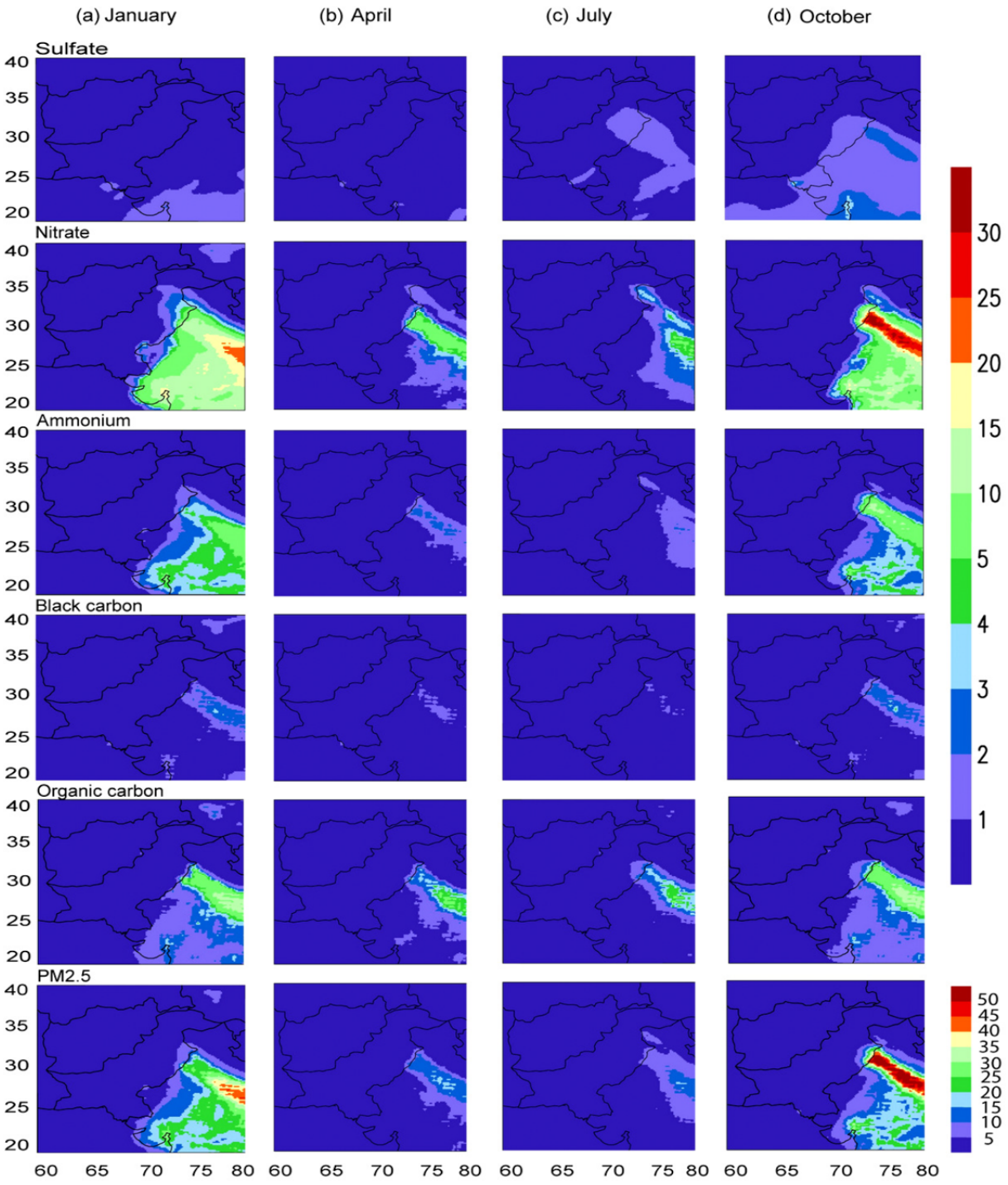

Fig. 15. Simulated monthly mean surface-layer concentrations (μg/m^3) of SO_4^{2-}, NO_3^-, NH_4^+, BC, OC, and $PM_{2.5}$ in the TransB simulation for (a) January, (b) April, (c) July, and (d) October of 2006.

India and Pakistan is found to be the major meteorological parameter that determines the transboundary aerosol transport. Comparisons our simulated aerosols with ground-based measurements of concentrations and satellite measurements of aerosol optical depth suggest further improvements in studies of aerosols in Pakistan. First, emission inventories of aerosol precursors and aerosols need continuing improvement in terms of both total amount and seasonal variations. With limited observational datasets available, the comparisons with observations at Lahore and Karachi show that the model can reproduce the magnitude of sulfate aerosol concentrations but tends to underestimate concentrations of NO_3^-, NH_4^+, OC, and $PM_{2.5}$. Second, nationwide long-term ground-based measurements of both aerosol precursors and aerosols are needed for evaluation of emission inventories and for improving the representations of chemical species in models. Third, natural aerosol species, such as mineral dust and sea salt, should be examined in

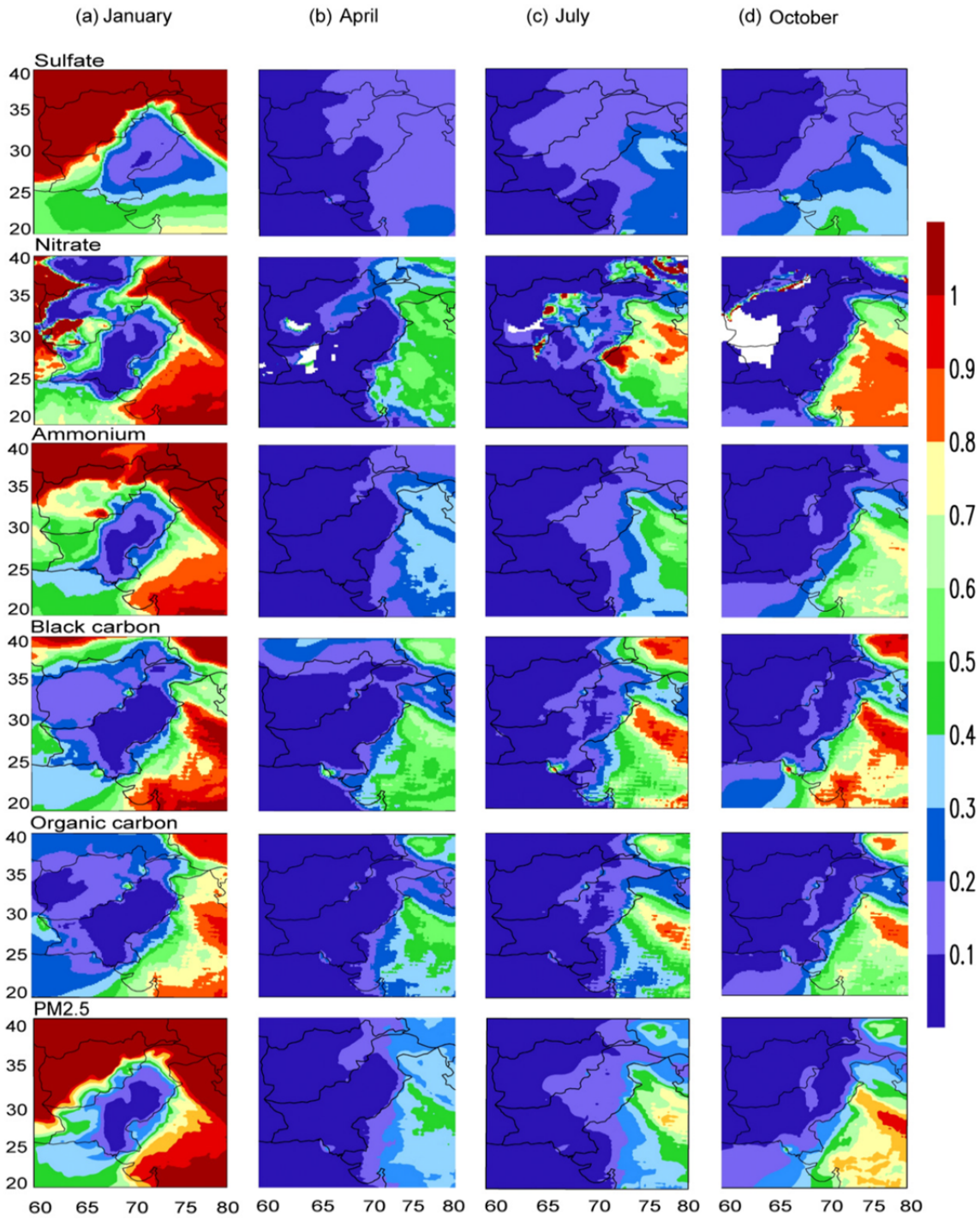

Fig. 16. Ratios of aerosol concentrations in TransB to those simulated in the CTRL simulation for (a) January, (b) April, (c) July, and (d) October of 2006.

our future work since these two species contribute largely to simulated and observed aerosol optical depth in April and July.

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