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Decadal trend and interannual variation of outflow of aerosols from East Asia: Roles of variations in meteorological parameters and emissions

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HIGHLIGHTS

- We quantify decadal and interannual variations of aerosol outflow from East Asia.
- Simulated annual outflow of $PM_{2.5}$ from East Asia increased 53% over 1986–2006.
- Outflow of $PM_{2.5}$ from East Asia had an average interannual variation of 5.2%.
- Decadal trends in aerosol outflow were mainly driven by changes in emissions.
- Interannual variations in aerosol outflow were driven by meteorological variables.

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ABSTRACT

We apply a global three-dimensional Goddard Earth Observing System (GEOS) chemical transport model (GEOS-Chem) to quantify the decadal trend and interannual variation of aerosol outflow from East Asia. Simulations of aerosols are performed for years 1986–2006, driven by the NASA/GEOS-4 assimilated meteorological fields. The impacts of variations in meteorological parameters and emissions are imposed separately and together by numerical experiments. With variations in both meteorological parameters and anthropogenic emissions, simulated annual outflow of $PM_{2.5}$ (sum of sulfate, nitrate, ammonium, black carbon, and organic carbon) from East Asia increased by 6.0 Tg (or 53%) over years 1986–2006, in which the outflow fluxes of sulfate, nitrate, and ammonium aerosols had the largest contributions with decadal trends of +1.2, +0.8, and +0.7 Tg decade^{-1}, respectively. Simulated outflow fluxes of aerosols also exhibited large interannual variations; the absolute percent departure from the mean (APDM) values of the annual outflow fluxes of sulfate, nitrate, ammonium, black carbon, organic carbon, and $PM_{2.5}$ were 5.9%, 7.9%, 5.4%, 5.8%, 4.2%, and 5.2%, respectively, as variations in both meteorological parameters and anthropogenic emissions were considered. Model sensitivity studies show that, for outflow fluxes of aerosols from East Asia, the decadal trends were driven by variations in anthropogenic emissions whereas the interannual variations were dominated by variations in meteorological parameters. The increases in anthropogenic emissions alone explained about 86% of the decadal trend in outflow of $PM_{2.5}$. Although variations in meteorological parameters can influence the interannual variations in aerosol outflow by changing both aerosol concentrations over East Asia and zonal winds, the latter is identified to be the key factor because of the high positive correlation coefficient between the annual outflow flux of $PM_{2.5}$ and zonal wind at 700 hPa (the altitude with the maximum outflow fluxes) for years 1986–2006.

1. Introduction

The outflow of aerosols from East Asia has drawn a lot of attention because of the relative high emissions and concentrations of aerosols in this region (Streets et al., 2003; Richter et al., 2005; Wang et al., 2006; Yu et al., 2012; Jiang et al., 2013; Luan and

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Jaegle, 2013; Matsui et al., 2013). Aerosols exported from East Asia have been shown to reach the North Pacific and North America through long-range transport (Richter et al., 2005; Chin et al., 2007; Fairlie et al., 2007; Liu et al., 2008). Park et al. (2004) reported that the transpacific transport of Asian pollutants contributed about 30% of the annual mean background sulfate in the United States based on the simulations using the GEOS-Chem model. Heald et al. (2006) showed that the transport from East Asia led to a seasonal mean increase in surface-layer sulfate concentration of 0.16 $\mu g\ m^{-3}$ in the northwestern United States in spring of 2001, by using satellite measurements of aerosol optical depth (AOD) over the North Pacific together with the GEOS-Chem simulations. Wang et al. (2009) found that the monthly mean background concentration of sulfate in the western United States increased by 0.4 $\mu g\ m^{-3}$ (20%) as a result of the emissions in Asia in April of year 2001, based on the simulations using the Community Multiscale Air Quality (CMAQ) model. With respect to other aerosol species, Hadley et al. (2007) estimated the long-range transport of black carbon across the Pacific Ocean into North America during April of 2004 using the Chemical Weather Forecast System (CFORS) model and found that over 75% of BC transported into North America originated from Asia. Matsui et al. (2013) simulated Asian BC outflow for years 2008–2010 using the CMAQ model and found that anthropogenic emissions from China, biomass burning emissions from Southeast Asia, and biomass burning emissions from Siberia and Kazakhstan contributed, respectively, 61%, 17%, and 6% to the total eastward BC flux averaged over the three years.

Previous studies reported that the outflow of aerosols from East Asia had strong seasonal variations, with maximum flux in spring and minimum value in summer when the seasonal changes in both meteorological parameters and emissions were considered (Yu et al., 2008, 2012; Matsui et al., 2013). In spring, warm conveyor belt occurs frequently over the Asian continent (Eckhardt et al., 2004), in which strong ascending airstreams lift pollutants into the free troposphere and then the westerlies carry them across the Pacific (Holzer et al., 2003, 2005; Wuebbles et al., 2007). The mid-latitude westerlies are the strongest in spring among all seasons, leading to the strongest outflow of aerosols in spring. The minimum outflow in summer results from the weak westerlies and the largest wet deposition of aerosols associated with the East Asian summer monsoon precipitation (Holzer et al., 2005; Matsui et al., 2013).

Few previous studies examined the interannual and decadal changes in aerosol outflow from East Asia. Industrial and biomass burning emissions in East Asia have large interannual variations (Carmichael et al., 2002; Duncan et al., 2003; Richter et al., 2005) and meteorological parameters associated with the Asian monsoon system also vary year by year (Huang et al., 2012), both of which can influence the interannual variations of the outflow of aerosols. Yu et al. (2008) estimated the outflow fluxes of aerosols over the western Pacific Ocean for years 2002–2005 using the Moderate Resolution Imaging Spectroradiometer (MODIS) AOD. They showed that the outflow was the strongest in year 2003, as a result of the large biomass burning emissions from spring to summer in Eurasia in that year. On decadal time scale, Jiang et al. (2013) investigated the projected 2000–2050 changes in concentrations of aerosols in China and the associated transboundary aerosol transport by using the GEOS-Chem model and reported that the annual outflow of $PM_{2.5}$ from eastern China was simulated to change by −7.0%, −0.7%, and −9.0% over 2000–2050 owing to climate change alone, changes in emissions alone, and changes in both climate and emissions, respectively, indicating that the meteorological parameters can be as important as emissions in influencing variations of outflow of aerosols from East Asia.

We present here a study to systematically examine the interannual variation and decadal trend of outflow of aerosols from East Asia, based on the simulations of aerosol mass fluxes for years 1986–2006 using the global chemical transport model GEOS-Chem. We aim to quantify (1) the interannual variation and decadal trend of aerosol outflow from East Asia for all major aerosol species (sulfate, nitrate, ammonium, black carbon, organic carbon) and $PM_{2.5}$ aerosol (sum of mass of all aerosol species), and (2) the roles of variations in meteorological parameters and/or emissions from anthropogenic and biomass burning sources in interannual variation and decadal trend of aerosol outflow from East Asia. The GEOS-Chem model and numerical experiments are described in Section 2. Section 3 presents simulated outflow fluxes of different aerosol species. Sections 4 and 5 examine, respectively, the decadal trends and interannual variations of outflow fluxes of aerosols from East Asia. The impacts of variations in biomass burning emissions on interannual variations of aerosol outflow fluxes are presented in Section 6. Section 7 discusses the uncertainties in model results that are associated with emissions inventories.

2. Model description and experimental design

2.1. GEOS-chem model

We simulate concentrations and transport of 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 horizontal resolution of 2° latitude by 2.5° longitude and 30 vertical layers from the surface to 0.01 hPa. The GEOS-Chem model includes a fully coupled treatment of tropospheric O_3-NO_x-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, 2004), mineral dust (Fairlie et al., 2007), and sea salt (Alexander et al., 2005). For transport of aerosols, the model uses the advection scheme of Lin and Rood (1996), the deep convective scheme of Zhang and McFarlane (1995), and the shallow convection scheme of Hack (1994).

The simulated aerosols in the GEOS-Chem model have been evaluated in a number of studies by using ground-based measurements in East Asia (Zhang et al., 2010; Fu et al., 2012; Jeong and Park, 2013; Jiang et al., 2013; Wang et al., 2013; Lou et al., 2014). Wang et al. (2013) found that simulated concentrations of sulfate, nitrate, and ammonium at 22 EANET sites in East Asia exhibited annual mean biases of −10%, +31%, and +35%, respectively. Fu et al. (2012) showed that the simulated annual mean concentrations of BC and OC averaged over rural and background sites were underestimated by 56% and 76%, respectively. Jiang et al. (2013) and Lou et al. (2014) reported similar magnitudes of biases in simulated aerosols in China in the GEOS-Chem model. The model, however, can capture the spatial distributions and seasonal variations of each aerosol species despite of the biases in simulated concentrations (Jiang et al., 2013). We will discuss how these model biases influence result of this work in Section 7.

2.2. Emissions

Global emissions of aerosols and their precursors in the GEOS-Chem model follow Park et al. (2003, 2004), with anthropogenic emissions of SO_2, NO_x, and NH_3 in Asia overwritten by the emissions inventories of Streets et al. (2003) for year 2000 and Zhang et al. (2009) for year 2006. Interannual variations in anthropogenic emissions are represented by annual scaling factors. To simulate anthropogenic emissions over 1986–2006, the scaling factors for SO_2 and NO_x follow those described in van Donkelaar

et al. (2008). For BC and OC, the scaling factors for years 1996–2006 are taken from Lu et al. (2011) and those for years 1986–1995 are derived from the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment Report (AR5) emissions inventories (Lamarque et al., 2010) using a linear interpolation approach. The IPCC AR5 inventories have emissions for every 10 years between 1850 and 2000. For NH_3, the scaling factors for years 1994–2005 are obtained from Dong et al. (2010) and those for years 1986–1993 are also derived from the IPCC AR5 emissions inventories. Biomass burning emissions are taken from the GFED-2 inventory for years 1997–2006 (van der Werf et al., 2006); we do not consider biomass burning emissions before 1997 because of the lack of datasets. Fig. 1 shows the variations in anthropogenic and biomass burning emissions of aerosols and aerosol precursors in Asia (60°–150°E, 10°S–56°N) over 1986–2006. Relative to 1986, year 2006 anthropogenic emissions of SO_2, NO_x, NH_3, BC, and OC increased by 99%, 188%, 42%, 40%, and 15%, respectively. During 1997–2006, biomass burning emissions of BC and OC exhibited larger interannual variations than anthropogenic emissions.

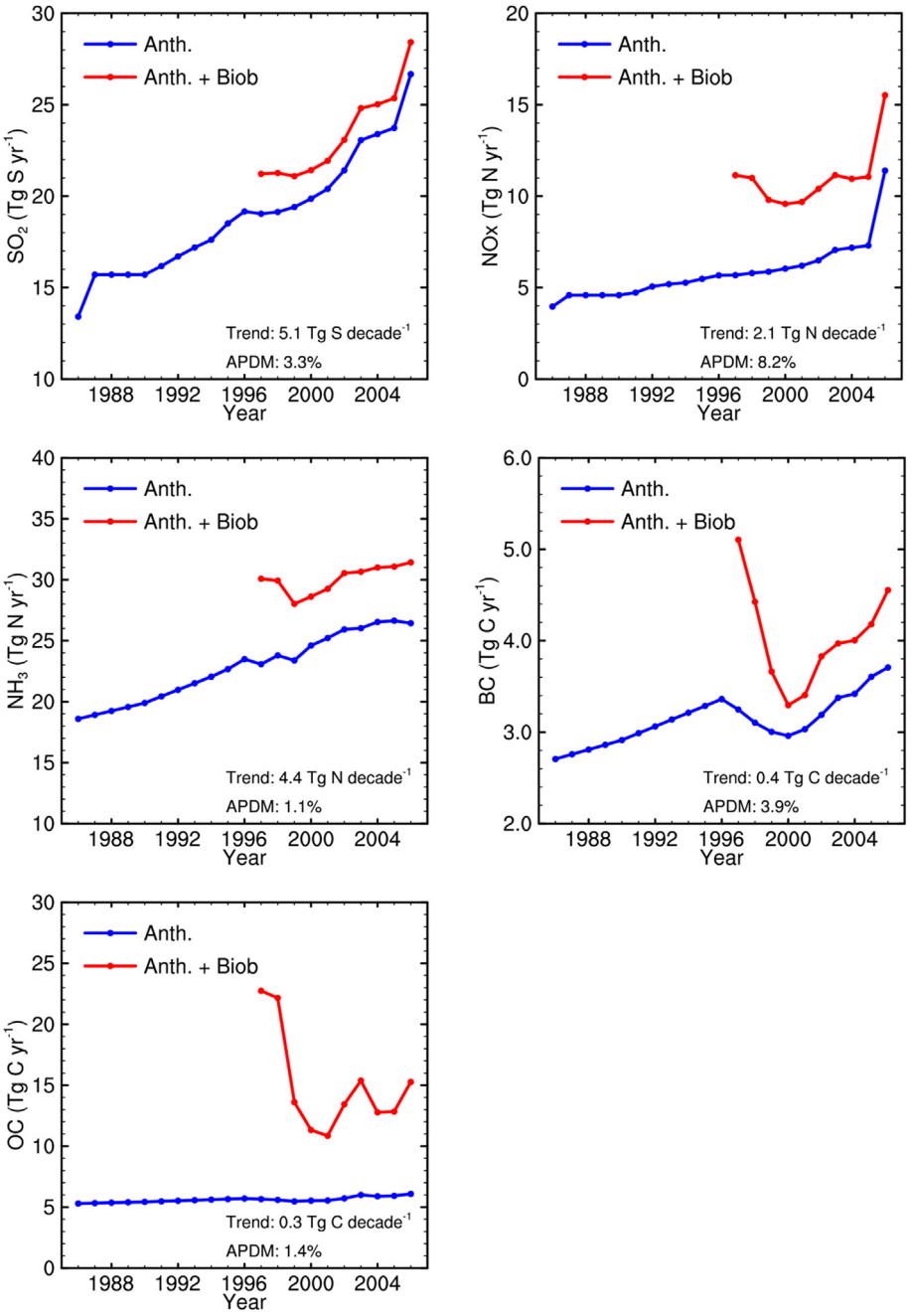

Fig. 1. Yearly variations in anthropogenic and biomass burning emissions of (a) SO_2, (b) NO_x, (c) NH_3, (d) BC, and (e) OC in Asia (60°–150°E, 10°S–56°N) over years 1986–2006. Blue lines are anthropogenic emissions and red lines represent total emissions (anthropogenic plus biomass burning emissions). The linear trend and absolute departure from the mean (APDM) of anthropogenic emissions are shown at the bottom right corner of each panel. Note that biomass burning emissions are not available before year 1997. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2.3. Experiments

The concentrations of aerosols and the associated aerosol outflow from East Asia are simulated for years 1986–2006, driven by the GEOS-4 meteorological fields. We perform the following simulations to identify the relative roles of variations in meteorological parameters and emissions in interannual variation and decadal trend of aerosol outflow from East Asia:

1) VALL: The simulation of aerosols and aerosol transport for years 1986–2006, with variations in both meteorological parameters and anthropogenic emissions over 1986–2006. Biomass burning emissions are not included in this simulation.
2) VMET: The simulation of aerosols and aerosol transport for years 1986–2006 to quantify the impact of variations in meteorological parameters alone on interannual variation and decadal trend of outflow of aerosols. Meteorological parameters are allowed to vary over 1986–2006. Anthropogenic emissions are fixed at year 2006 level. Biomass burning emissions are not included in this simulation.
3) VEMIS: The simulation of aerosols and aerosol transport for years 1986–2006 to quantify the impact of variations in anthropogenic emissions alone on interannual variation and decadal trend of outflow of aerosols. The meteorological parameters are fixed at year 2006 values. Anthropogenic emissions are allowed to vary over 1986–2006. Biomass burning emissions are not included in this simulation.
4) VALLB: The simulation of aerosols and aerosol transport for years 1997–2006 with variations in meteorological parameters, anthropogenic emissions, and biomass burning emissions. The model setups are the same as those in VALL for years 1997–2006 except that biomass burning emissions are considered and are allowed to vary over 1997–2006.

The outflow fluxes of aerosols from East Asia are calculated as the mass fluxes through the vertical plane from the surface to 100 hPa altitude along 135°E from 20° to 60°N.

3. Simulated outflow fluxes of different aerosol species

Fig. 2 shows the monthly outflow fluxes of different aerosol species from simulation VALL that are averaged over years 1986–2006. All aerosol species exhibited maximum outflow in spring and minimum flux in August. Sulfate aerosol had the largest annual outflow flux, followed by NO_3^-, NH_4^+, OC, and BC; the annual outflow fluxes of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC from East Asia contributed 7.4 Tg yr^{-1} (53%), 1.7 Tg yr^{-1} (12%), 2.5 Tg yr^{-1} (18%), 0.7 Tg yr^{-1} (5%), and 1.7 Tg yr^{-1} (12%), respectively, to the 14.0 Tg yr^{-1} of annual outflow of $PM_{2.5}$ (sum of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC aerosols).

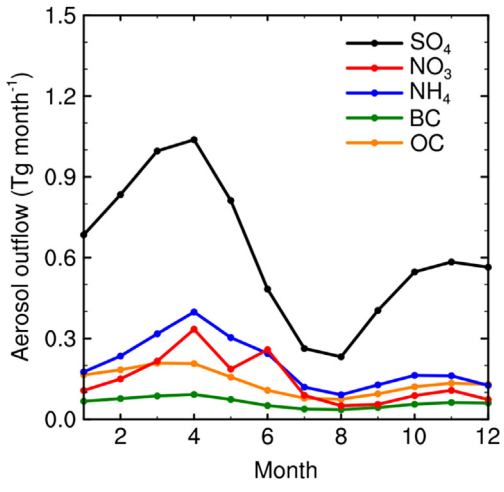

Fig. 2. Monthly outflow fluxes of different aerosol species from simulation VALL that are averaged over years 1986–2006.

Fig. 3 shows the simulated seasonal outflow fluxes of $PM_{2.5}$ across the selected plane along 135°E from 20° to 60°N that are averaged over years 1986–2006 of simulation VALL. Simulated $PM_{2.5}$ mass fluxes were the largest in March–April–May (MAM) and the lowest in June–July–August (JJA), in agreement with the seasonal variation reported in Yu et al. (2008, 2012) and Matsui et al. (2013). Such seasonal variation in outflow flux was mainly driven by seasonal changes in meteorological parameters (Holzer et al., 2005; Yu et al., 2008). The maximum outflow flux of $PM_{2.5}$ was located between 850 and 500 hPa, over 30°N in DJF and over about 40°N in other seasons. Averaged over 1998–2006, the outflow fluxes of $PM_{2.5}$ from East Asia were 3.6, 5.4, 2.2, and 2.7 Tg $season^{-1}$ in December–January–February (DJF), MAM, JJA, and September–October–November (SON), respectively. Our estimates agree closely with the fluxes of 4.0, 6.8, 2.2, and 3.0 Tg $season^{-1}$ in these seasons derived from the MODIS AOD for year 2004 in Yu et al. (2008).

Previous studies have evaluated the magnitude and seasonal variation of Asian aerosol outflow simulated in the GEOS-Chem model. Park et al. (2005) showed that the GEOS-Chem model could reproduce the concentrations and vertical profiles of sulfate, nitrate, and BC aerosols over the Asian Pacific Rim in March–April of 2001 during the NASA Transport and Chemical Evolution over the Pacific (TRACE-P) aircraft campaign. Luan and Jaegle (2013) compared the GEOS-Chem simulated seasonal mean AOD with MODIS AOD averaged over years 2004–2010 in the North Pacific Ocean, and found that the model captured well the magnitude of AOD, with low biases of less than 20% in SON, DJF, and MAM and low biases of 20–40 % in JJA. For the purpose of this work, we compare in Fig. 4 the simulated interannual variations in annual mean AOD in simulation VALLB with those obtained from MODIS. Both the simulated and observed AOD values are the averages over the selected region (30°–50°N, 130°–150°E) in the western Pacific Ocean. The MODIS datasets are available since year 2001, and simulation VALLB with variations in meteorological parameters as well as anthropogenic and biomass burning emissions are available for 1997–2006 (the GEOS-4 meteorological fields are available for 1986–2006 and biomass burning emissions are not available before 1997). Over 2001–2006, the model captures the peaks and troughs of the MODIS AOD, with a high correlation coefficient of +0.66. Sulfate and mineral dust aerosols are the most dominant contributors to simulated AOD in the Asian outflow region (Luan and Jaegle, 2013). By simulating AOD of mineral dust and sulfate aerosols in the GEOS-Chem model for years 2004–2010, Luan and Jaegle (2013) showed that, while dust AOD generally exceeded sulfate AOD in spring of every year, sulfate AOD was larger than dust AOD on an annual mean basis.

4. Simulated decadal trends in outflow fluxes of aerosols

4.1. Effect of variations in both meteorological parameters and anthropogenic emissions

Fig. 5a shows the annual aerosol outflow fluxes from East Asia for years 1986–2006 simulated in VALL simulation. The outflow fluxes of all aerosol species exhibited an increasing trend over 1986–2006; relative to year 1986, the annual outflow fluxes of

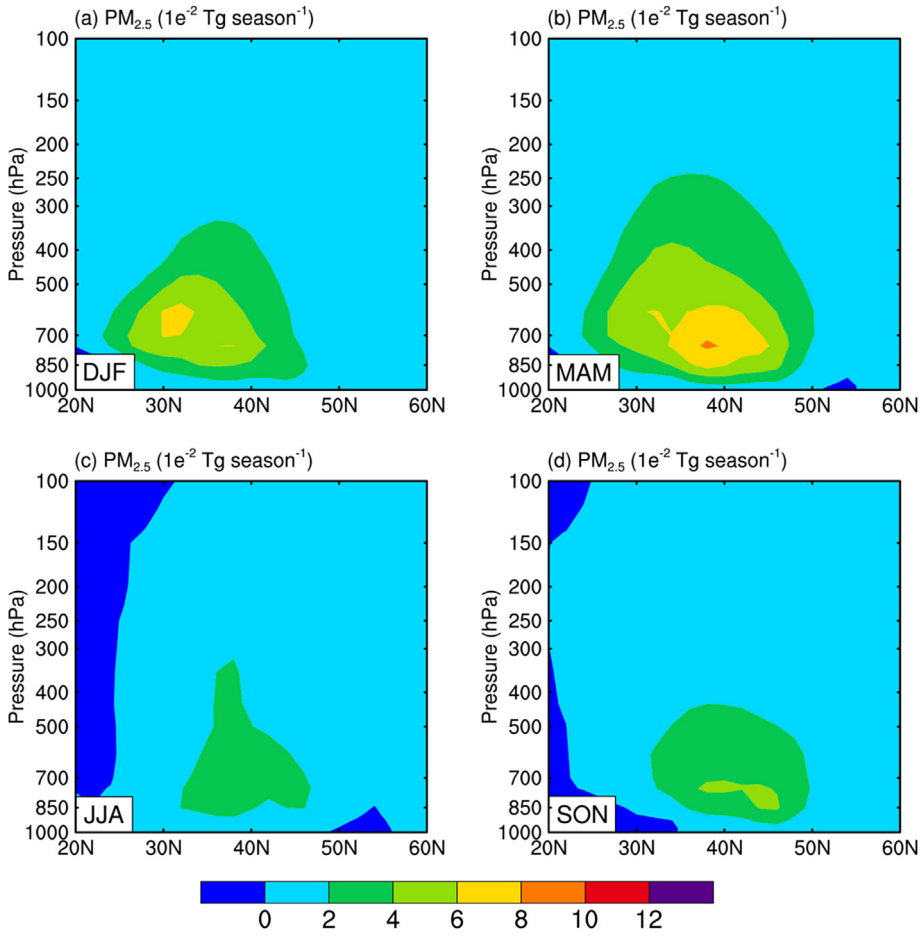

Fig. 3. Simulated seasonal outflow fluxes of $PM_{2.5}$ across the selected plane along 135°E from 20° to 60°N that are averaged over years 1986–2006 of simulation VALL. Positive values represent eastward fluxes and negative values represent westward fluxes.

SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC in year 2006 increased by 47%, 118%, 72%, 50%, and 20%, respectively. As a result, the annual outflow of $PM_{2.5}$ increased from 11.3 Tg in 1986 to 17.3 Tg in 2006, which was an increase of 6.0 Tg (53%) over 1986–2006.

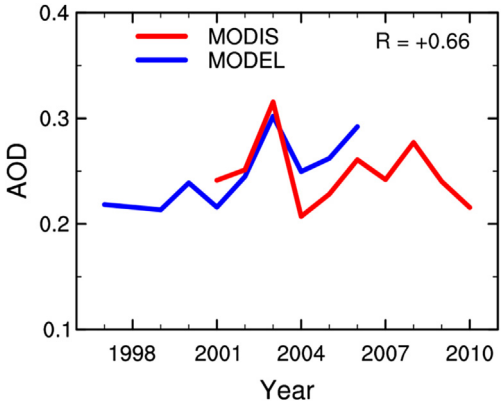

Fig. 4. Comparison simulated aerosol optical depth (AOD) with MODIS AOD. Both the simulated and observed AOD values are the averages over the selected region (30°–50°N, 130°–150°E) in the western Pacific Ocean. The MODIS datasets are available since year 2001, and modeled AOD values from simulation VALLB (with variations in meteorological parameters as well as anthropogenic and biomass burning emissions) are available for 1997–2006. Correlation coefficient R between simulations and observations is calculated over the time period of 2001–2006.

The decadal trends in outflow fluxes of aerosols for the studied period are calculated using the least square fit and summarized in Table 1. With variations in both meteorological parameters and anthropogenic emissions in VALL simulation, the decadal trends in outflow fluxes of SO_4^{2-}, NO_3^-, NH_4^+, BC, OC, and $PM_{2.5}$ were respectively, +1.2, +0.8, +0.7, +0.1, +0.1, and +2.8 Tg decade^{-1}, indicating that the increase in outflow of $PM_{2.5}$ over the 21-yr period resulted mainly from the increases in outflow of sulfate, nitrate, and ammonium aerosols. Although OC has a larger contribution to the outflow of $PM_{2.5}$ than BC, the decadal trend in outflow flux of OC is the same as that of BC, because of the small increase (15%) in anthropogenic OC emission over 1986–2006 as shown in Fig. 1. It should be noted that the decadal trends were different in different seasons (Table 1). While the simulated outflow of $PM_{2.5}$ in MAM accounted for 55% of the annual outflow of $PM_{2.5}$ averaged over 1986–2006, the decadal trend in seasonal outflow of $PM_{2.5}$ in MAM was +1.2 Tg decade^{-1}, which contributed 43% of the decadal increasing trend in annual outflow flux of $PM_{2.5}$.

Fig. 5b shows the deviation from the mean in aerosol outflow flux obtained in simulation VALL for 1986–2006. The deviation from the mean is defined as $DM_i = (P_i - 1/n\sum_{i=1}^{n}P_i)/1/n\sum_{i=1}^{n}P_i$, where P_i is the simulated outflow flux of an aerosol in year i, and n is the number of years examined ($n = 21$ for years 1986–2006). From 1986 to 2006, the values of DM for outflow fluxes of all aerosol species generally changed from negative values to positive values, which also indicated an increasing trend in aerosol outflow from East Asia over 1986–2006. The outflow flux of NO_3^- exhibited the

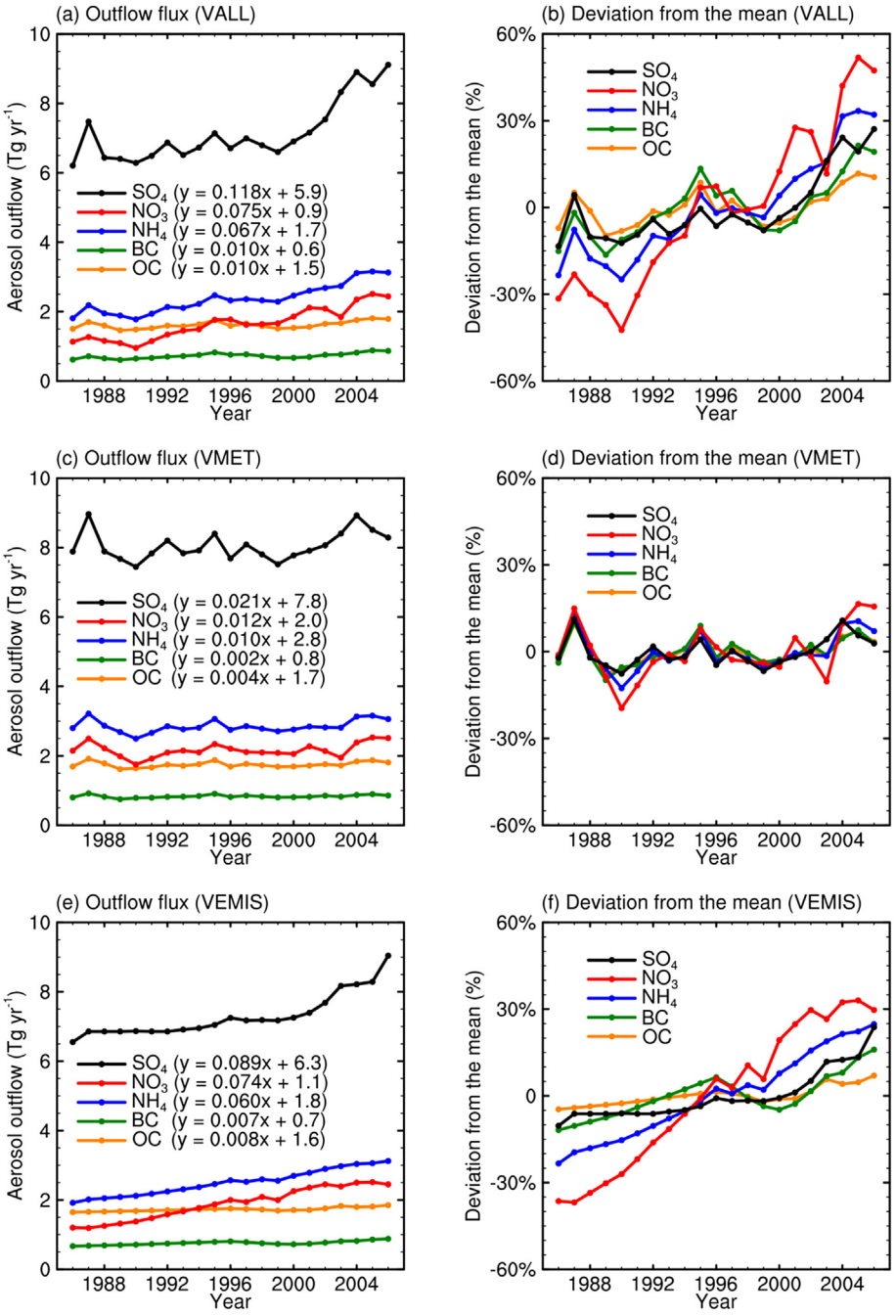

Fig. 5. Annual aerosol outflow fluxes from East Asia for years 1986–2006 simulated in simulations (a) VALL, (c) VMET, and (e) VEMIS with the associated deviation from the mean in (b), (d) and (f), respectively. The decadal trends in outflow fluxes are calculated using the least square fit approach and are presented in parenthesis in (a), (c), and (e). See text for the definition of deviation from the mean.

largest variations in DM value; the DM value changed from −31.5% in 1986 to +47.4% in 2006.

4.2. *Effect of variations in meteorological parameters alone*

Fig. 5c presents the outflow fluxes of aerosols from East Asia for years 1986–2006 obtained in VMET simulation and Fig. 5d shows the associated DM values. With changes in meteorological parameters alone, the outflow fluxes of aerosols showed very small increasing trends over years 1986–2006, with the decadal trends obtained from the linear square fit much smaller than those found in VALL simulation. For example, the annual outflow flux of $PM_{2.5}$ increased by 1.2 Tg decade^{-1} from 1986 to 2006 in VMET simulation, which accounted for only 20% of the simulated increase in $PM_{2.5}$ outflow of 6.0 Tg decade^{-1} over the same time period in VALL simulation, indicating that the variations in meteorological parameters were not the key factors that led to the decadal trends in outflow of aerosols from East Asia. The DM values of outflow fluxes of aerosols from simulation VMET also showed very small increasing trends over years 1986–2006 (Fig. 5d).

Table 1
Simulated seasonal and annual outflow fluxes of aerosols in years 1986 and 2006 from simulations VALL, VMET, and VEMIS. Percentage changes in outflow fluxes of aerosols [(flux in 2006 − flux in 1986)/(flux in 1986) × 100%] are shown in brackets. The decadal trends in outflow fluxes of aerosols are obtained by the least square fit over years 1986–2006. The unit is Tg for outflow fluxes and Tg decade^{-1} for decadal trends in outflow fluxes. The outflow fluxes of aerosols from East Asia are calculated as the mass fluxes through the vertical plane from the surface to 100 hPa altitude along 135°E from 20° to 60°N.

Species	Season	VALL			VMET			VEMIS		
		Outflow in 1986	Outflow in 2006 (Percentage change)	Decadal trend	Outflow in 1986	Outflow in 2006 (Percentage change)	Decadal trend	Outflow in 1986	Outflow in 2006 (Percentage change)	Decadal trend
SO_4^{2-}	DJF	2.0	2.4 (20%)	0.28	2.4	2.3 (−4%)	0.03	1.8	2.2 (22%)	0.21
	MAM	2.5	3.7 (48%)	0.50	3.2	3.4 (6%)	0.12	2.6	3.7 (42%)	0.37
	JJA	0.7	1.3 (86%)	0.21	1.0	1.2 (20%)	0.01	0.8	1.3 (63%)	0.20
	SON	1.4	1.7 (21%)	0.14	1.6	1.6 (0%)	0.04	1.4	1.7 (21%)	0.07
	ANN	6.2	9.1 (47%)	1.18	7.9	8.3 (5%)	0.21	6.6	9.0 (36%)	0.89
NO_3^-	DJF	0.2	0.4 (100%)	0.12	0.4	0.4 (0%)	0.02	0.3	0.5 (67%)	0.12
	MAM	0.5	1.1 (120%)	0.34	1.0	1.1 (10%)	0.06	0.5	1.1 (120%)	0.34
	JJA	0.2	0.6 (200%)	0.17	0.4	0.6 (50%)	0.03	0.3	0.6 (100%)	0.16
	SON	0.2	0.3 (50%)	0.12	0.3	0.4 (33%)	0.01	0.1	0.3 (200%)	0.11
	ANN	1.1	2.4 (118%)	0.75	2.2	2.5 (14%)	0.12	1.2	2.5 (108%)	0.74
NH_4^+	DJF	0.5	0.6 (20%)	0.11	0.6	0.6 (0%)	0.01	0.4	0.6 (50%)	0.10
	MAM	0.8	1.4 (75%)	0.31	1.3	1.4 (8%)	0.06	0.8	1.4 (75%)	0.28
	JJA	0.3	0.6 (100%)	0.14	0.5	0.6 (20%)	0.01	0.3	0.6 (100%)	0.13
	SON	0.4	0.5 (25%)	0.11	0.5	0.5 (0%)	0.02	0.3	0.5 (67%)	0.09
	ANN	1.8	3.1 (72%)	0.67	2.8	3.1 (11%)	0.10	1.9	3.1 (63%)	0.60
BC	DJF	0.2	0.2 (0%)	0.02	0.2	0.2 (0%)	0.00	0.2	0.2 (0%)	0.01
	MAM	0.2	0.3 (50%)	0.04	0.3	0.3 (0%)	0.02	0.2	0.3 (50%)	0.03
	JJA	0.1	0.2 (100%)	0.02	0.1	0.1 (0%)	0.00	0.1	0.1 (0%)	0.01
	SON	0.1	0.2 (100%)	0.02	0.2	0.2 (0%)	0.00	0.1	0.2 (100%)	0.01
	ANN	0.6	0.9 (50%)	0.10	0.8	0.9 (13%)	0.02	0.7	0.9 (29%)	0.07
OC	DJF	0.5	0.5 (0%)	0.00	0.5	0.5 (0%)	0.00	0.5	0.5 (0%)	0.02
	MAM	0.5	0.7 (40%)	0.05	0.6	0.7 (17%)	0.02	0.6	0.7 (17%)	0.03
	JJA	0.2	0.3 (50%)	0.03	0.2	0.3 (50%)	0.01	0.2	0.3 (50%)	0.01
	SON	0.3	0.4 (33%)	0.02	0.4	0.4 (0%)	0.01	0.3	0.4 (33%)	0.02
	ANN	1.5	1.8 (20%)	0.10	1.7	1.8 (6%)	0.04	1.6	1.8 (13%)	0.08
$PM_{2.5}$	DJF	3.3	4.0 (21%)	0.54	4.3	4.1 (−5%)	0.05	3.2	4.0 (25%)	0.46
	MAM	4.6	7.1 (54%)	1.23	6.4	6.8 (6%)	0.28	4.8	7.1 (48%)	1.05
	JJA	1.4	2.9 (107%)	0.56	2.2	2.7 (23%)	0.06	1.7	2.9 (71%)	0.52
	SON	2.4	3.1 (29%)	0.41	3.0	3.0 (0%)	0.08	2.4	3.1 (29%)	0.31
	ANN	11.3	17.3 (53%)	2.80	15.3	16.5 (8%)	0.49	12.0	17.3 (44%)	2.38

4.3. *Effect of variations in anthropogenic emissions alone*

Simulation VEMIS with changes in anthropogenic emissions alone reproduces a large fraction of the decadal increase in aerosol outflow obtained in VALL (Fig. 5e). Using the linear fit, the decadal trends in annual outflow fluxes of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC in VEMIS simulation were, respectively, +0.9, +0.7, +0.6, +0.1, and +0.1 Tg decade^{-1}. Therefore, the decadal changes in anthropogenic emissions were the major drivers of the decadal increases in aerosol outflow fluxes from East Asia.

In VEMIS simulation, ratios of outflow fluxes of SO_4^{2-}, NO_3^-, NH_4^+, BC, and OC to outflow of $PM_{2.5}$ changed, respectively, from 55.0%, 10.0%, 15.8%, 5.8%, and 13.3% in year 1986 to 52.0%, 14.5%, 17.9%, 5.2%, and 10.4% in year 2006. From 1986 to 2006, the ratios of the outflow fluxes of NO_3^- and NH_4^+ to the outflow of $PM_{2.5}$ increased whereas the ratios of the outflow fluxes of SO_4^{2-}, BC, and OC decreased, indicating the increasing importance of NO_3^- and NH_4^+ aerosols in outflow of aerosols from East Asia.

5. Simulated interannual variations in outflow fluxes of aerosols

To analyze the interannual variations in aerosol outflow fluxes, the decadal trends in simulated outflow fluxes are firstly identified by linear square fit (as shown in Section 4) and removed from the time series, following the approach used in previous studies that examined interannual variations in ozone concentrations (Camp et al., 2003), sea surface temperature, partial pressure of CO_2 (Gruber et al., 2002), sea level pressure (Thompson and Wallace, 1998), and North Atlantic Oscillation index (Jung et al., 2003). Then the interannual variations in outflow fluxes are quantified by mean absolute deviation (MAD) and absolute percent departure from the mean (APDM) defined as $MAD = 1/n\sum_{i=1}^{n}|P_i - 1/n\sum_{i=1}^{n}P_i|$ and $APDM = 100\% \times MAD/(1/n\sum_{i=1}^{n}P_i)$, where P_i is the detrended outflow flux of an aerosol in year i, and n is the number of years examined (n = 21 for years 1986–2006). Therefore MAD represents the absolute interannual variation averaged over the n years and APDM represents the interannual variation in percentage relative to the average outflow over the n years.

Fig. 6 shows the deviations of annual outflow fluxes of aerosols from the 21-yr (1986–2006) mean as the decadal trends are removed from the time series from each of the simulations VALL, VMET, and VEMIS. While the deviations were in the range of ±20% in simulations VALL and VMET, the values were within ±12% in simulation VEMIS. Note that the peaks and troughs in deviations in VALL simulation were similar to those in VMET in terms of magnitude and the year of occurrence, suggesting that the interannual variations in aerosol outflow fluxes were dependent on variations in meteorological parameters instead of the variations in anthropogenic emissions. With variations in meteorological parameters alone in simulation VMET, the deviations of outflow fluxes of BC and OC showed peaks and troughs in the same years over 1986–2006. Black carbon, as a chemically inert tracer, responded to changes in meteorological parameters interannually. Outflow fluxes of SO_4^{2-}, NO_3^-, and NH_4^+ exhibited larger deviations than BC and reached their peaks and troughs in different years from BC, resulting from the chemical reactions influenced by variations in

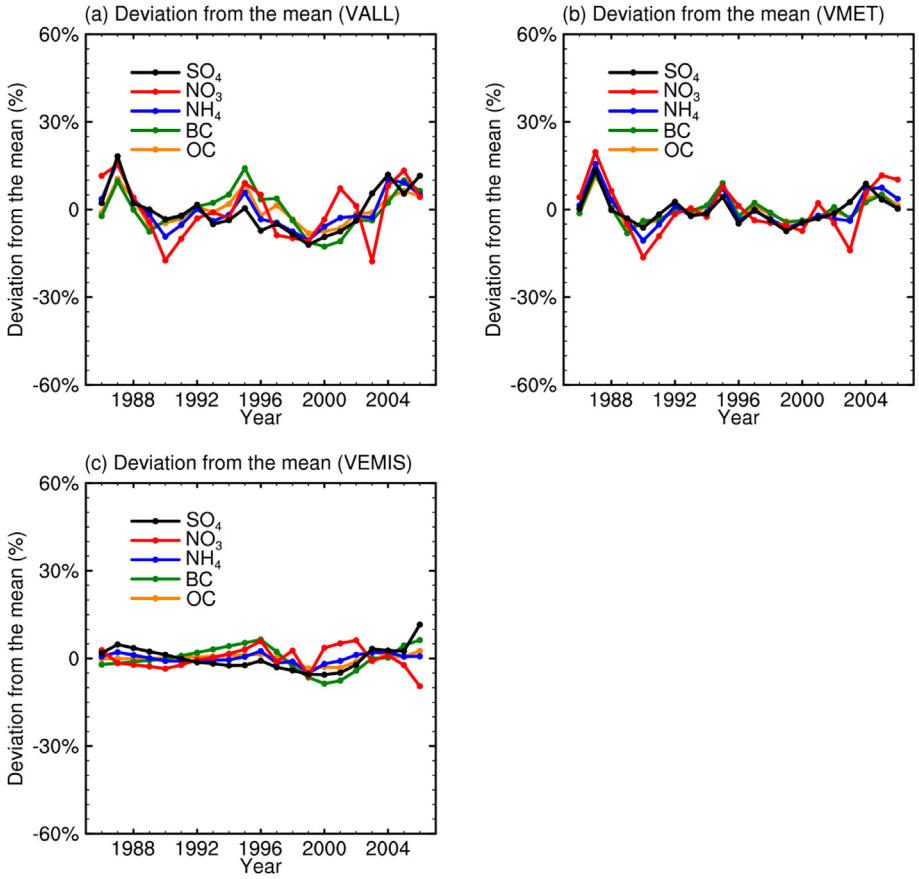

Fig. 6. Deviation from the mean of the detrended outflow fluxes of different aerosol species from simulations (a) VALL, (b) VMET, and (c) VEMIS for 1986–2006.

meteorological parameters. For example, in VMET simulation, the outflow fluxes of SO_4^{2-}, NO_3^-, and NH_4^+ exhibited troughs in year 1990, but the outflow BC did not have a trough in this year.

Fig. 7 summarizes the APDM values of the detrended outflow fluxes of aerosols from the VALL, VMET, and VEMIS simulations. In VALL simulation, seasonal outflow fluxes of SO_4^{2-}, NH_4^+, BC, and OC showed similar magnitude of interannual variation with APDM values of 5–9%, and these APDM values were generally larger in MAM and JJA than in SON and DJF. For NO_3^-, the APDM values of the outflow fluxes were the largest among all aerosol species with seasonal APDM values of 8–15%, and the maximum APDM occurred in DJF. As a result of the interannual variations of individual aerosol species, the APDM values of the seasonal outflow fluxes of $PM_{2.5}$ were 5–8% and the APDM value of the annual outflow fluxes of $PM_{2.5}$ was 5.2% in VALL simulation. With variations in meteorological parameters alone in VMET simulation, the APDM values were very close to those in VALL simulation, indicating again that the interannual variations in aerosol outflow fluxes were mainly driven by variations in meteorological parameters. Variations in meteorological parameters influence interannual variations in outflow fluxes of aerosols through the following ways. First, variations in meteorological parameters change aerosol concentrations over East Asia (Jeong and Park, 2013; Mu and Liao, 2014). Second, variations in meteorological parameters influence outflow of aerosols by changing transport. The simulated outflow flux of $PM_{2.5}$ is found to correlate positively with zonal wind at 700 hPa (the altitude with the maximum outflow fluxes) averaged over 20°–60°N along 135°E, with a high correlation coefficient of +0.76 for years 1986–2006, indicating that the variation in zonal wind is the key factor that led to interannual variations in outflow fluxes of aerosols in VMET.

Relative to the VMET simulation, the APDM values of outflow fluxes of aerosols in VEMIS were much smaller; the APDM values of seasonal outflow fluxes of $PM_{2.5}$ from the VEMIS simulation were 1–2%. In VEMIS, the APDM values of annual outflow fluxes of SO_4^{2-}, NO_3^-, and BC were in the range of 3–4%, larger than the values of 1–2% for NH_4^+ and OC, corresponding to the relatively larger APDM values of 3–8% in annual anthropogenic emissions of SO_2, NO_x, and BC whereas smaller APDM values of 1–2% in anthropogenic emissions of NH_3 and OC (Fig. 1).

6. Effect of variability in biomass burning emissions

Because of the large interannual variations in biomass burning emissions (Fig. 1), we show in Table 2 the MAD and APDM values of outflow fluxes of aerosols for years 1997–2006 in VALL and VALLB simulations. Interannual variations in biomass burning emissions over 1997–2006 are found to lead to large interannual variation in outflow of OC; the MAD (APDM) value of the outflow of OC from East Asia was 0.05 Tg yr^{-1} (3.3%) in simulation VALL and 0.48 Tg yr^{-1} (14.8%) in simulation VALLB. This mainly resulted from the large biomass burning emissions of OC over South and Southeast Asia and in Russia in spring and summer (Giglio et al., 2006; Bian et al., 2007). For outflow fluxes of other aerosol species, MAD (APDM) values in VALLB became slight smaller than those in VALL, which can be explained by that the interannual variations in biomass burning were out of phase with those caused by meteorological fields and anthropogenic emissions. Despite of the large interannual variation in outflow of OC driven by biomass burning

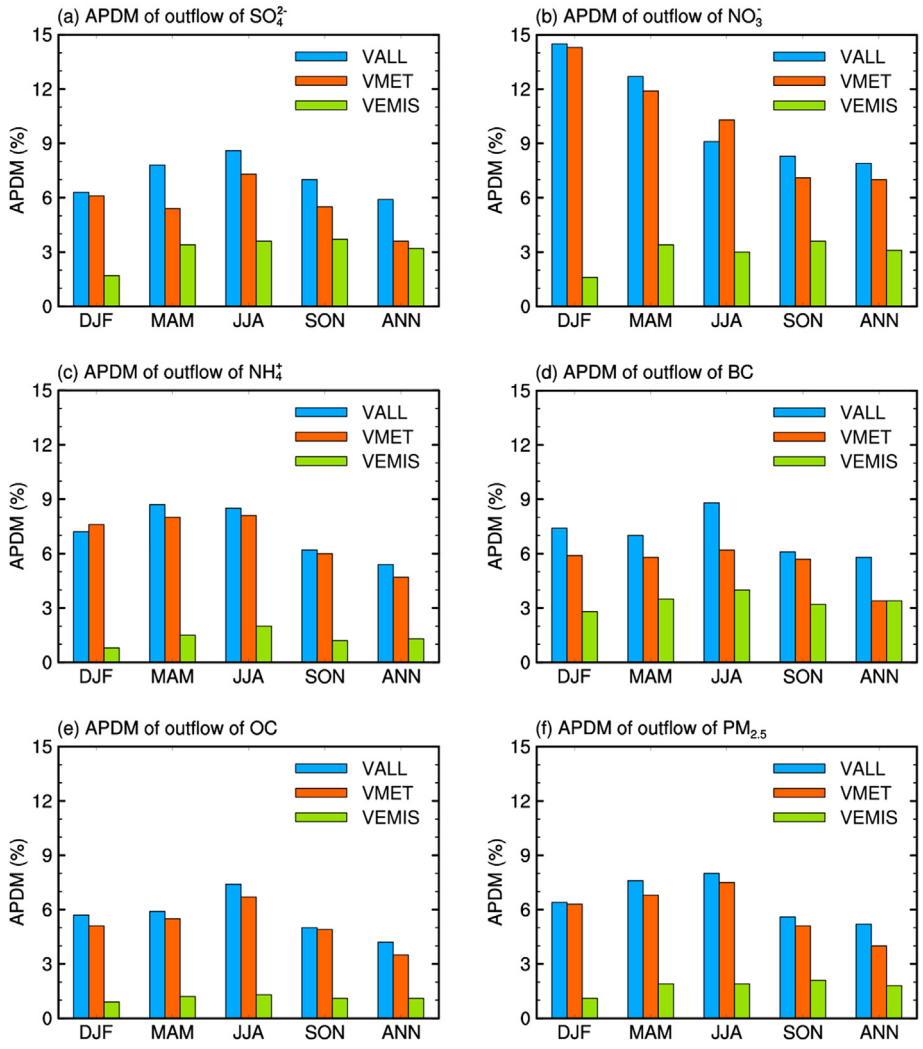

Fig. 7. The APDM values of the detrended seasonal and annual outflow fluxes of (a) SO_4^{2-}, (b) NO_3^-, (c) NH_4^+, (d) BC, (e) OC, and (f) $PM_{2.5}$ from simulations VALL, VMET, and VEMIS for 1986–2006.

emissions, the MAD (APDM) value of the outflow of $PM_{2.5}$ exhibited a small change from 0.47 Tg yr^{-1} (3.2%) in VALL simulation to 0.65 Tg yr^{-1} (3.8%) in VALLB simulation.

7. Uncertainties associated with emissions inventories

There are some sources of uncertainties in our simulations that need to be improved in future studies. The anthropogenic emissions inventories are the key sources of uncertainties for estimated aerosol outflow fluxes from East Asia. The evaluations of simulated aerosols in the GEOS-Chem model in the studies of Fu et al. (2012), Jiang et al. (2013), and Lou et al. (2014) have suggested that emissions of carbonaceous aerosols are currently underestimated in China. By using measurements reported in the literature in recent years, Fu et al. (2012) showed that the simulated annual mean concentrations of BC and OC averaged over rural and background sites were underestimated by 56% and 76%, respectively. Jiang et al. (2013) and Lou et al. (2014) reported similar magnitudes of biases in simulated carbonaceous aerosols in China in the GEOS-Chem model. Underestimation of BC in China was also found in all Aerosol Comparisons between Observations and Models (AeroCom) models (Koch et al., 2009). Therefore, our study may have underestimated the contributions of BC and OC to aerosol outflow from East Asia.

We also compare in Fig. 8 and Table 3 the anthropogenic emissions used in this work with other emissions inventories that have multiple years of emissions available. We consider emission inventories of Regional Emission inventory in Asia (REAS) v1.1 for 1986–2003 (Ohara et al., 2007), REAS v2.1 for 2000–2006 (Kurokawa et al., 2013), Emissions Database for Global Atmospheric Research (EDGAR)-Hemispheric Transport of Air Pollution (HTAP) v1 for 2000–2005 (Janssens-Maenhout et al., 2012), and EDGAR v4.2 for 1986–2006 (http://edgar.jrc.ec.europa.eu). Anthropogenic emissions of SO_2, NO_x, NH_3, BC, and OC from all inventories show approximately linear increasing trends, except for those of SO_2 in EDGAR v4.2. The uncertainties in Asian anthropogenic emissions are not expected to influence our results about the interannual variations of aerosol outflow fluxes (i.e., interannual variations in aerosol outflow are mainly driven by meteorological variables), considering that the decadal trends in simulated outflow fluxes are firstly identified by linear square fit and removed from the time series (Section 5).

Table 3 compares the decadal trends in anthropogenic emissions from different emissions inventories. The decadal trends are calculated only for the inventories with datasets longer than 10

Table 2
Simulated mean absolute deviation (MAD) and absolute percent departure from the mean (APDM) of the detrended aerosol outflow fluxes from East Asia for years 1997–2006. The unit is Tg yr^{-1} for MAD and percentage for APDM. See definitions of MAD and APDM in the text.

Species	MAD (APDM)	
	VALL	VALLB
SO_4^{2-}	0.30 (3.9%)	0.27 (3.4%)
NO_3^-	0.11 (5.5%)	0.06 (2.8%)
NH_4^+	0.09 (3.2%)	0.06 (2.0%)
BC	0.05 (6.0%)	0.04 (4.2%)
OC	0.05 (3.3%)	0.48 (14.8%)
$PM_{2.5}$	0.47 (3.2%)	0.65 (3.8%)

years (i.e., decadal trends are calculated for REAS v1.1 with datasets over 1986–2003 and EDGAR v4.2 with datasets over 1986–2006). The decadal trend of SO_2 emission is +5.1 Tg S $decade^{-1}$ in our study, which is about the same as the trend of +5.4 Tg S $decade^{-1}$ in the REAS v1.1 inventory but larger than that of +3.5 Tg S $decade^{-1}$ in the inventory of EDGAR v4.2. For NO_x emission, the decadal trends are similar in this work, REAS v1.1, and EDGAR v4.2, with values of +2.1, +2.8, and +2.7 Tg N $decade^{-1}$, respectively. For NH_3 emission, the decadal trend is +4.4 Tg N $decade^{-1}$ in this work, slightly larger than the value of 3.7 Tg N $decade^{-1}$ in EDGAR v4.2. The BC emissions used in our work agree with those in the REAS v1.1 in that they both exhibit increases before 1996, decreases over 1996–2000, and then increases again after 2000 (Fig. 8). Therefore,

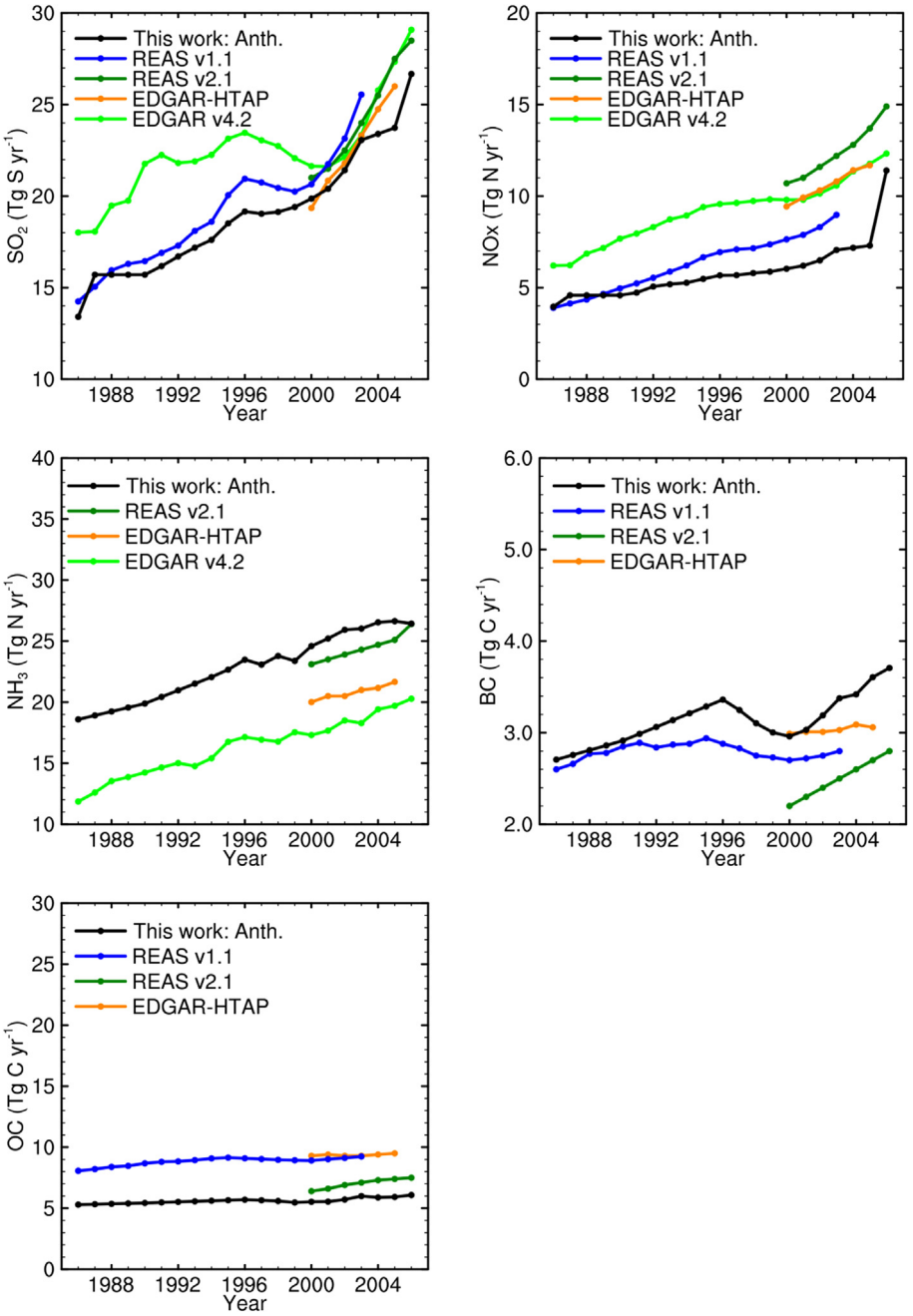

Fig. 8. Comparisons of the yearly variations in regional total anthropogenic emissions used in this work with those from other emissions inventories for (a) SO_2, (b) NO_x, (c) NH_3, (d) BC, and (e) OC. See Table 3 for the detailed information of these emissions inventories.

Table 3
Comparison of the mean values and decadal trends of anthropogenic emissions used in this work with those from other emissions inventories. Mean values are obtained by averaging the regional total emissions over years with datasets available. The decadal trends are calculated only for the inventories with datasets longer than 10 years.

Emissions	Species	Time	Region	Mean value	Decadal trend	Reference
This work	SO_2	1986–2006	60°–150°E, 10°S–56°N	18.9 Tg S yr^{-1}	5.1 Tg S $decade^{-1}$	
	NO_x			5.8 Tg N yr^{-1}	2.1 Tg N $decade^{-1}$	
	NH_3			22.8 Tg N yr^{-1}	4.4 Tg N $decade^{-1}$	
	BC			3.1 Tg C yr^{-1}	0.4 Tg C $decade^{-1}$	
	OC			5.6 Tg C yr^{-1}	0.3 Tg C $decade^{-1}$	
REAS v1.1	SO_2	1986–2003	Asian domain	19.0 Tg S yr^{-1}	5.4 Tg S $decade^{-1}$	Ohara et al. (2007)
	NO_x			6.2 Tg N yr^{-1}	2.8 Tg N $decade^{-1}$	
	BC			2.8 Tg C yr^{-1}	0.1 Tg C $decade^{-1}$	
	OC			8.8 Tg C yr^{-1}	0.5 Tg C $decade^{-1}$	
EDGAR v4.2	SO_2	1986–2006	60°–150°E, 10°S–56°N	22.4 Tg S yr^{-1}	3.5 Tg S $decade^{-1}$	http://edgar.jrc.ec.europa.eu
	NO_x			9.1 Tg N yr^{-1}	2.7 Tg N $decade^{-1}$	
	NH_3			16.3 Tg N yr^{-1}	3.7 Tg N $decade^{-1}$	
REAS v2.1	SO_2	2000–2006	Asian domain	24.4 Tg S yr^{-1}		Kurokawa et al. (2013)
	NO_x			12.4 Tg N yr^{-1}		
	NH_3			24.4 Tg N yr^{-1}		
	BC			2.5 Tg C yr^{-1}		
	OC			7.0 Tg C yr^{-1}		
EDGAR-HTAP v1	SO_2	2000–2005	60°–150°E, 10°S–56°N	22.8 Tg S yr^{-1}		Janssens-Maenhout et al. (2012)
	NO_x			10.6 Tg N yr^{-1}		
	NH_3			20.8 Tg N yr^{-1}		
	BC			3.0 Tg C yr^{-1}		
	OC			9.4 Tg C yr^{-1}		

the decadal trends of emissions in this study agree, to some extent, with those in other emissions inventories. Since the decadal trends in aerosol outflow flux are mainly driven by changes in emissions, further studies are needed as improved long–term emissions of aerosol and aerosol precursors in East Asia are available for the calculation of outflow fluxes of aerosols.

8. Conclusions

We examine the impacts of variations in meteorological parameters and emissions on decadal trend and interannual variation of outflow of aerosols (sulfate, nitrate, ammonium, black carbon, organic carbon, and $PM_{2.5}$) from East Asia on the basis of the GEOS-Chem simulations of aerosols over years 1986–2006 driven by the assimilated GEOS-4 meteorological fields. The impacts of variations in meteorological parameters and emissions are imposed separately and together by sensitivity studies.

Model simulation shows the annual outflow of $PM_{2.5}$ from East Asia increased from 11.3 Tg in 1986 to 17.3 Tg in 2006, which was an increase of 53% over 1986–2006. The decadal trends in outflow fluxes of SO_4^{2-}, NO_3^-, NH_4^+, BC, OC, and $PM_{2.5}$ were, respectively, +1.2, +0.8, +0.7, +0.1, +0.1, and +2.8 Tg $decade^{-1}$, indicating that the increase in outflow of $PM_{2.5}$ over the 21-yr period resulted mainly from the increases in outflow fluxes of sulfate, nitrate, and ammonium aerosols. Sensitivity studies indicate that the increases in anthropogenic emissions alone explained about 86% of the decadal trend in outflow of $PM_{2.5}$ simulated with variations in both meteorological parameters and anthropogenic emissions.

The interannual variations in outflow fluxes of aerosols are quantified by mean absolute deviation (MAD) and absolute percent departure from the mean (APDM) values after removing the linear trend in aerosol outflow fluxes over 1986–2006. With variations in both meteorological parameters and emissions, the seasonal outflow fluxes of SO_4^{2-}, NH_4^+, BC, and OC showed similar magnitude of interannual variations with APDM values of 5–9%, and the outflow fluxes of NO_3^- exhibited the largest interannual variation among all aerosol species with APDM values of 8–15%. As a result, the APDM values of the seasonal outflow fluxes of $PM_{2.5}$ were 5–8% and the APDM value of the annual outflow fluxes of $PM_{2.5}$ was 5.2% in VALL simulation. Sensitivity simulations show that the interannual variations in aerosol outflow were dominated by variations in meteorological parameters instead of those in anthropogenic emissions. Variations in meteorological parameters drove the interannual variations in aerosol outflow by changing aerosol concentrations over East Asia and by changing the zonal winds, and the latter was the key factor since the outflow flux of $PM_{2.5}$ correlated with zonal wind at 700 hPa (the altitude with the maximum outflow fluxes) averaged over 20°–60°N along 135°E with a high correlation coefficient of +0.76 for years 1986–2006.

We also perform a sensitivity simulation to examine the impacts of biomass burning emissions on interannual variation of outflow of aerosols from East Asia. Interannual variations in biomass burning emissions over 1997–2006 are found to lead to large interannual variation in outflow of OC; the APDM value of the outflow of OC from East Asia was 3.3% in simulation VALL and 14.8% in simulation VALLB. However, biomass burning emissions are simulated to have a small contribution to interannual variations in outflow of $PM_{2.5}$ from East Asia.

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