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THE RADIATIVE EFFECTS OF ANTHROPOGENIC AEROSOLS OVER CHINA AND THEIR SENSITIVITY TO SOURCE EMISSION

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Abstract: In this paper, the RIEMS 2.0 model, source emission in 2006 and 2010 are used to simulate the distributions and radiative effects of different anthropogenic aerosols over China. The comparison between the results forced by source emissions in 2006 and 2010 also reveals the sensitivity of the radiative effects to source emission. The results are shown as follows: (1) Compared with those in 2006, the annual average surface concentration of sulfate in 2010 decreased over central and eastern China with a range of −5 to 0 μg/m^3; the decrease of annual average aerosol optical depth of sulfate over East China varied from 0.04 to 0.08; the annual average surface concentrations of BC, OC and nitrate increased over central and eastern China with maximums of 10.90, 11.52 and 12.50 μg/m^3, respectively; the annual aerosol optical depths of BC, OC and nitrate increased over some areas of East China with extremes of 0.006, 0.007 and 0.008, respectively. (2) For the regional average results in 2010, the radiative forcings of sulfate, BC, OC, nitrate and their total net radiative forcing at the top of the atmosphere over central and eastern China were −0.64, 0.29, −0.41, −0.33 and −1.1 W/m^2, respectively. Compared with those in 2006, the radiative forcings of BC and OC in 2010 were both enhanced, while that of sulfate and the net radiative forcing were both weakened over East China mostly. (3) The reduction of the cooling effect of sulfate in 2010 produced a warmer surface air temperature over central and eastern China; the maximum value was 0.25 K. The cooling effect of nitrate was also slightly weakened. The warming effect of BC was enhanced over most of the areas in China, while the cooling effect of OC was enhanced over the similar area, particularly the area between Yangtze and Huanghe Rivers. The net radiative effect of the four anthropogenic aerosols generated the annual average reduction and the maximum reduction were -0.096 and −0.285 K, respectively, for the surface temperature in 2006, while in 2010 they were −0.063 and −0.256 K, respectively. In summary, the change in source emission lowered the cooling effect of anthropogenic aerosols, mainly because of the enhanced warming effect of BC and weakened cooling effect of scattering aerosols.

Key words: aerosol radiative effect; radiative forcings; sulfate; nitrate; organic carbon; black carbon
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1 INTRODUCTION

Atmospheric aerosols can be divided into anthropogenic aerosols and natural aerosols according to their different sources. Anthropogenic aerosols play an important role in the impact of human activities on regional-to-global-scale climate. Wide attention has been paid to the effects of anthropogenic aerosols in the field of Atmospheric Science and Environmental Science. The Fifth Assessment Report (AR5) of the Intergovernmental Panel on Climate Change shows that aerosols have a high degree of inhomogeneity in time and space and their distributions vary largely among different regions, which makes it difficult to study their impacts on climate (Collins et al.[1]). Thus, more intensive studies on climatic impact of aerosols especially anthropogenic aerosols are necessary.

In recent years, many researchers have carried out a considerable number of studies on anthropogenic aerosols. Sun and Liu [2, 3] studied the impact of sulfate and BC on the East Asian summer monsoon using CAM3.0 model and they found that sulfate contributed to the cooling in most areas of China, reduction of precipitation and the weakening of East Asian summer monsoon. They also demonstrated that BC weakened the East Asian summer monsoon and their relationship was far more complex. Menon et al.[4] investigated the connection between BC and regional precipitation and they thought the emission of BC was associated with the trend of precipitation in China in recent decades, increasing summer flooding in southern China and drought in northern China. The results of Jacobson and Kaufman [5] show that BC may reduce the wind speed. The regional air quality model system was also served

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as tools to study the distribution of carbon aerosols over China in summer and the results show that OC mainly accumulates in central and eastern regions, as well as some regions of northern China (Han et al.[6]). In addition, the emission of BC was found to bring about cooling in northern China and warming in southern China in a study using the regional climate model named RegCM3 (Zhang and Yin[7]). Han et al.[8] found that the combined direct radiative effects of anthropogenic aerosols and natural aerosols over East Asia showed typical seasonal variations.

The IPCC report pointed out that the impact of aerosol emissions and their scavenging in the atmosphere on climate change needs further studies [1]. Since the climate effects of aerosols lie on their concentration simulation, emission source then makes a significant contribution to the uncertainty in the climate effects of aerosols. Additionally, anthropogenic aerosols such as sulfate and nitrate are usually generated by their gaseous precursors through photochemical process and their concentrations are related to meteorological conditions. Hence, there is a nonlinear relationship between the change of emission source and the variation of aerosol concentration. Besides, emission inventories contain large uncertainties (Liao et al.[9]). With the rapid development of the economy and the implementation of energy-saving and emission-reduction policies, the emissions of aerosols and their precursors as well as the emission ratios of different species are changing at the same time. The IPCC also reported that after the reduction of emissions, secondary aerosols such as nitrate still have large uncertainty [1]. As a result, it is necessary to update source emission in the model on time, evaluate the effects of anthropogenic aerosols and then judge how the climate effects of anthropogenic aerosols change after emission reduction.

For the past few years, the emission amount and structure of anthropogenic aerosols have been changing all the time because of China's rapid economic development and enhanced human activities. Since coal use is one of the major anthropogenic sources of black carbon (Penner et al.[10]) and coal plays a dominant role in the energy-consuming of China, it is not surprising that one-fourth of anthropogenic global BC is emitted there (Cooke et al.[11]; Streets et al.[12]). These years, with the rapid economic development in China, concentrations of air pollutants are very high [9] and emission of carbon aerosols in China has caught pervasive attention. In 2011, United Nations Environment Programme (UNEP), together with World Meteorological Organization, specially listed the emissions reduction scenario of BC in China in a comprehensive assessment report on BC and tropospheric ozone to emphasize the importance of BC (Emberson et al.[13]). "The Union of Climate and Clean Air" was established in the United States in February 2012 for the sake of stressing the importance of short-lived species such as BC (Wang and Zhou[14]). At the same time, a energy-saving and emission-reduction policy was put forward during "the Eleventh Five-Year Plan" in China. Since China is facing large pressure on climate diplomacy, it urgently needs to calculate the radiative forcings of anthropogenic aerosols at present in China and the changes in the climate effects of each aerosol, especially BC, after the implementation of the "energy-saving and emission-reduction" policy.

Typically, the aforementioned studies aiming at analyzing the effects of anthropogenic aerosols using regional models were forced by emission sources in 2006 or even before 2006, while a few used new emission sources. Besides, some studies (Han et al.[15]) on aerosols over East Asian areas recently were not focused on evaluating the sensitivity of aerosol climate effects to source emissions. In this paper, the RIEMS 2.0 model, source emission in 2006 and high resolution source emission in 2010 are used to compare the climate effects of sulfate, nitrate, BC and OC in China and discuss the climate effects sensitivities of anthropogenic aerosols to different emission sources.

2 MODEL INTRODUCTION AND EXPERIMENT DESIGN

2.1 *Mesoscale numerical model RIEMS2.0*

The climate model used in this study was the Regional Integrated Environmental Model System which was developed by the Key Laboratory of Regional Climate-Environment for Temperate East Asia (RCE-TEA), Chinese Academy of Sciences based on RIEMS1.0 version (Fu et al.[16]).

The chemical transport module and aerosol module are coupled with the RIEMS2.0 (Liu et al [17]). The aerosol module involves sulfate, nitrate, black carbon, organic carbon, secondary organic carbon, dust and sea salt aerosols, among which BC, dust sea salt aerosols and primary OC are primary aerosols. For the primary aerosols, only the emission, transport, diffusion and dry (wet) deposition processes are considered, but the chemical process is excluded. However, for secondary aerosols, the chemical process is very important. The chemistry model involves the CBM4 chemical mechanism (Carbon Bond Mechanism) (Gery et al.[18]) with 36 species and 83 chemical reactions included. In order to compute sulfate, nitrate and secondary organic aerosols, the thermodynamics equilibrium model ISORROPIA (CMAXS) (Hildemann et al.[19]) and the secondary organic model, which is from the air-quality model REMSAD (Regional Model System for Aerosols and Deposition) developed by U.S Environment Protection Agency (Odum et al. [20]; Griffin et al. [21]), are used in the RIEMS2.0.

Three mixture states of sulfate BC and nitrate are considered in this model and they are external mixture, internal mixture partly and internal mixture completely. In this paper, an external mixture state between

aerosols is assumed. This model has been used to simulate regional climates in East Asia (Fu et al.[22]; Zhang et al.[23]).

2.2 *Introduction of data*

The longitude and latitude for the center of the simulated domain are 32° N and 107° E, respectively. The model simulates a grid domain of 55 by 65 with the horizontal grid size of 60 km. There are 16 vertical layers and the top layer is at 100 hPa. NCEP/NCAR reanalysis data from 2006 at six-hour intervals were used as boundary conditions and the initial field for the climate model calculation in this study.

The emission inventory in 2006 and that in 2010 were used in this paper. The emission inventory in 2006 came from the Asian emission data of 2006 put forward by Zhang et al.[24] in the NASA's INTEX-B Plan with the resolution of 0.5° by 0.5°. The emission inventory in 2010 with the resolution of 0.25° by 0.25° was the MEIC-2010 provided by professor Zhang of Tsinghua University. Thirty-two kinds of species such as SO_2, BC, OC, VOCs are included in the source inventory.

2.3 *Numerical experiments design*

Twelve numerical experiments were conducted in this study and the conditions of each experiment are shown in Table 1. Aerosol species refer to the aerosols considered in each experiment. As is known, the emission inventory only contains the gaseous precursors such as SO_2 and NO_x while we need to consider the climate effect of sulfate and nitrate finally. In fact, most of sulfate was from the secondary aerosol computation in the model and we took 1% of the mass of SO_2 emission as another small fraction. The nitrate in this experiment was all computed by the secondary aerosol computation model. The radiative effects of aerosols were computed by utilizing the radiation scheme twice sequentially in the model. The first pass included all kinds of the aerosols while the second pass excluded one of the four aerosols and the radiative effect of this aerosol can then be obtained by the difference between the results of above two passes. The radiative effects of BC, sulfate, nitrate and OC in 2006 were computed from the difference between the results of case01 and that from case02 to case05, respectively. So were those in 2010. The net effect of these four kinds of anthropogenic aerosols in 2006 was obtained from the difference between case01 and case06 while in 2010 that was computed by case07 versus case12.

Table 1. Introduction of numerical experiments.

Experiment	Emission source	Aerosol species
case01/case07	2006/2010	Sulfate, nitrate, BC, OC
case02/caseo8	2006/2010	Sulfate, nitrate, OC
case03/case09	2006/2010	Nitrate, BC, OC
case04/case10	2006/2010	Sulfate, BC, OC
case05/case11	2006/2010	Sulfate, nitrate, BC
case06/case12	2006/2010	none

The emissions in western areas are much less than those in eastern areas relatively, which induces the low concentrations of aerosols in the west. As such, we mainly analyze aerosols and their climate effects in the central and eastern China, which is also called analysis domain in the later text. Fig.1 displays the whole research area and the area has been divided into four different domains to discuss.

The different domains defined in this paper are as follows: central and eastern China (22.58°N to 44.30°N, 101.14°E to 124.33°E); East China (27.84°N to 38.74°N, 111.53°E to 124.33°E); South China (22.58°N to 27.84° N, 101.14° E to 124.33° E); North China (38.74° N to 44.30°N, 101.14°E to 124.33°E); Central China (27.84° N to 38.74°N, 101.14°E to 111.53°E).

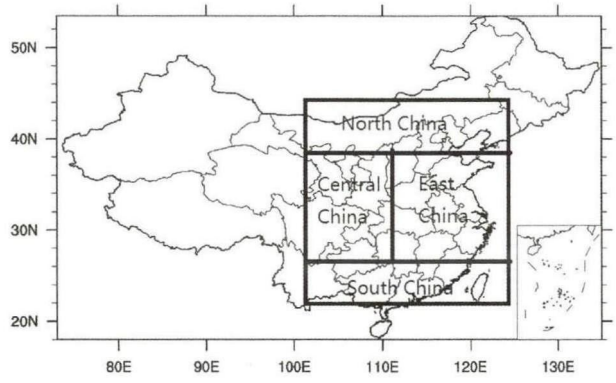

Figure 1. Different domains defined over eastern and central China.

3 RESULTS AND DISCUSSION

3.1 *Distributions of aerosol emissions*

The emissions of the four species in 2010 and their differences between 2010 and 2006 are given in Fig.2 and Fig.3, respectively. As can be seen from Fig.3, the change in emission inventory brought in the variations of the emission distributions of the four species. Compared to that in 2006, the emission of SO_2 in 2010 increased obviously over the eastern part of Sichuan Basin, as well as North China and a few developed regions over East China, with the maximum of 7.41g/ (s km^2). While over most of other areas of Central and eastern China, the emission of SO_2 decreased or presented a minor change. Furthermore, from Table 3, the emission of SO_2 decreased over all regions on average except North China. These are consistent with the findings of the study (Chen and Li[25]) that pollution control has been strengthened in our country and great progress has been made in pollution prevention in recent years. The emission of BC increased in some regions of South China and North China. In the regions where the economy is rapidly developing such as Pearl River Delta (PRD) region, the emission of BC increased with the maximum of 0.19g/ (s km^2). In stark contrast, the emission of BC in other areas in the southeast coast

of the Chinese mainland decreased. PRD is a typical urban agglomeration region with high commercialization and industrialization. Its increasing car numbers (Liao et al.[26]) contributes to its carbon emission, which is in agreement with another study (Yu[27]). In regard to OC, often co-emitted with BC, its emissions increased in most regions of North China and some regions of East China and PRD region, with the extreme of 0.16 g/(s km^2) in Wuhan. As for nitrogen oxides, their emissions mainly increased in North China and some economically developed regions over East China, with the maximum of 3.56 g/(s km^2).While respectively speaking, the distribution of the changes in nitrogen oxides emissions was spatially uniform and the high values were decentralized.

On the whole, compared to those in 2006, SO_2 e-

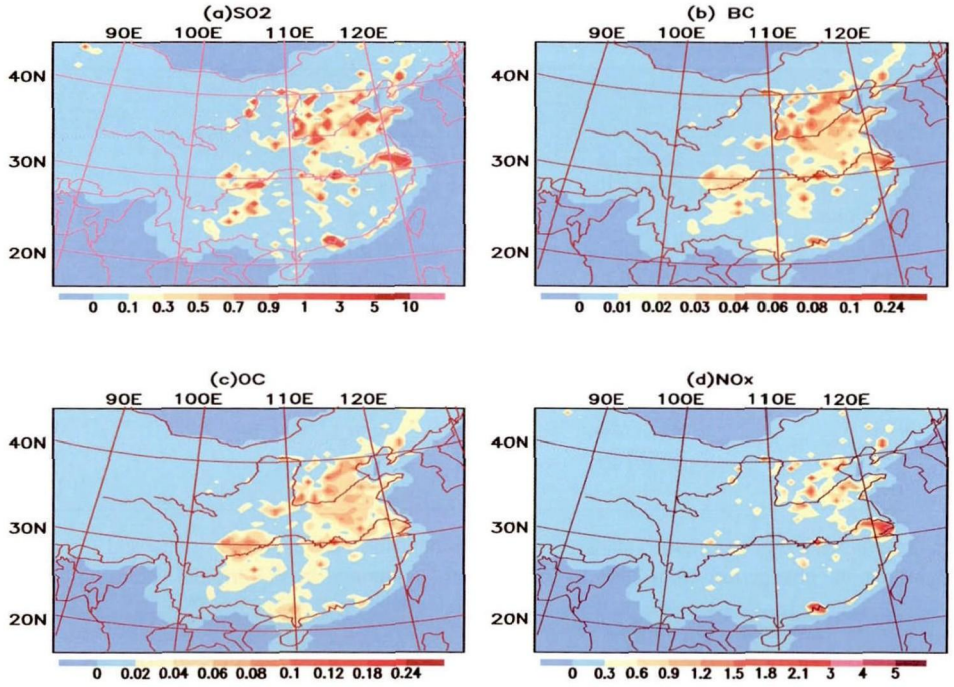

Figure 2. Distributions of the gaseous emissions in 2010. (a) SO_2 ; (b) BC; (c) OC; (d) NO_x. Units: g/(s km^2).

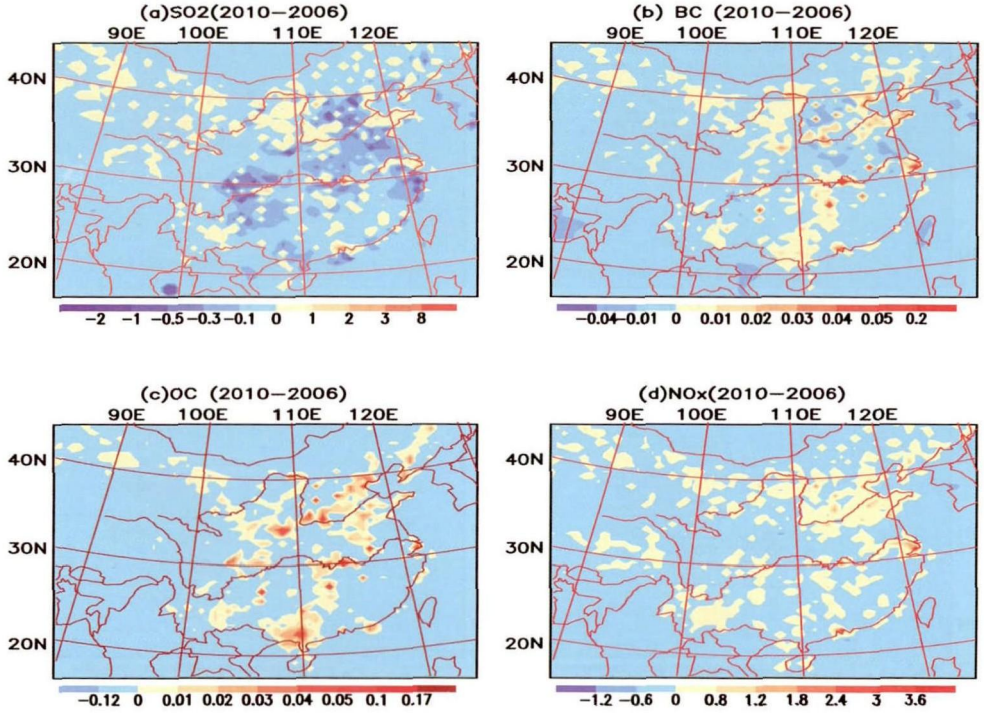

Figure 3. Distributions of the biases between two emission sources. (a) SO_2 ; (b) BC; (c) OC; (d) NO_x. Units: g/(s km^2).

missions in 2010 broadly decreased in central and eastern China while emissions of BC, OC and NO_x in 2010, especially NO_x, increased in many regions over central and eastern China and decreased in some other regions. Also evident is a pronounced increase of SO_2, BC, OC and NO_x in the regions with rapid economic development such as Beijing-Tianjin-Hebei, Yangtze River delta (YRD) and PRD. It is worth noticing that as shown in statistical results, the average emissions of BC and OC decreased subtly almost over each domain, whereas the decrease in the emission of SO_2 was manifest in central and eastern China generally (Tables 2 and 3). Yet, the average emissions of NO_x increased over all the domains, which was contrast to other species.

Table 2. Emissions of aerosols and their precursors and the regional average concentration of aerosols over central and eastern China.

Aerosols	Year	Emissions(Gg year^{-1})	Average column burdens (mg m^{-2})	Average surface concentrations (μg m^{-3})
Sulfate	2010	25491.58	2.68 (12.43)	2.22 (71.61)
	2006	28534.49	5.60 (17.29)	1.96 (9.59)
	2010/2006	0.89	0.48	1.13
BC	2010	1554.83	0.33 (1.84)	0.87 (12.34)
	2006	1627.33	0.23 (0.66)	0.48 (2.21)
	2010/2006	0.96	1.42	1.82
OC	2010	2913.87	1.86 (3.58)	2.65 (12.21)
	2006	3216.16	1.81 (3.14)	2.40 (5.40)
	2010/2006	0.91	1.03	1.11
Nitrate	2010	24347.06	1.34 (2.95)	0.49 (12.96)
	2006	18211.94	1.36 (1.92)	0.41 (1.87)
	2010/2006	1.34	0.99	1.21

Notes: The values in brackets are the maximum values of concentrations; the precursors of sulfate and nitrate are SO_2 and NO_x. (2010-2006)/2006 represents the change rate from 2006 to 2010 and it is computed by the original values without approximation.

3.2 *Distributions of aerosol concentrations*

The surface concentrations of the four aerosols are shown in Fig.4 and their differences between 2006 and 2010 can be seen in Fig.5. Overall, distributions of surface concentrations of aerosols are in good consistency with the distributions of their precursor emissions.

The surface concentration of sulfate in 2010 basically decreased in a range of 0 to 5 μg m^{-3} over many

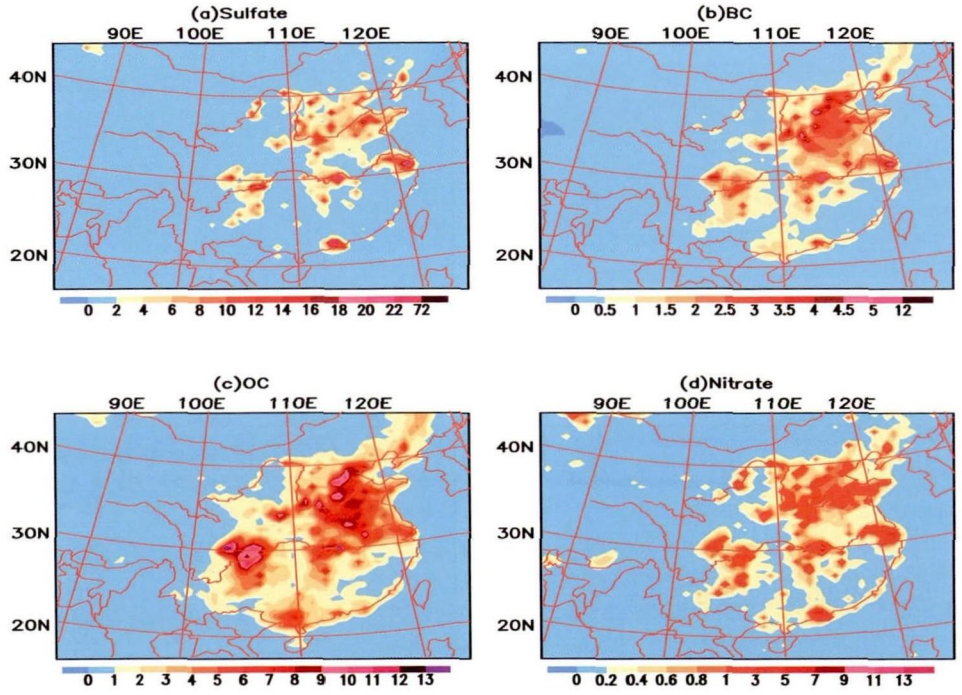

Figure 4. Distributions of the annual mean surface concentrations of aerosols in 2010. (a) Sulfate; (b) BC; (c) OC; (d) Nitrate. Units: μg m^{-3}.

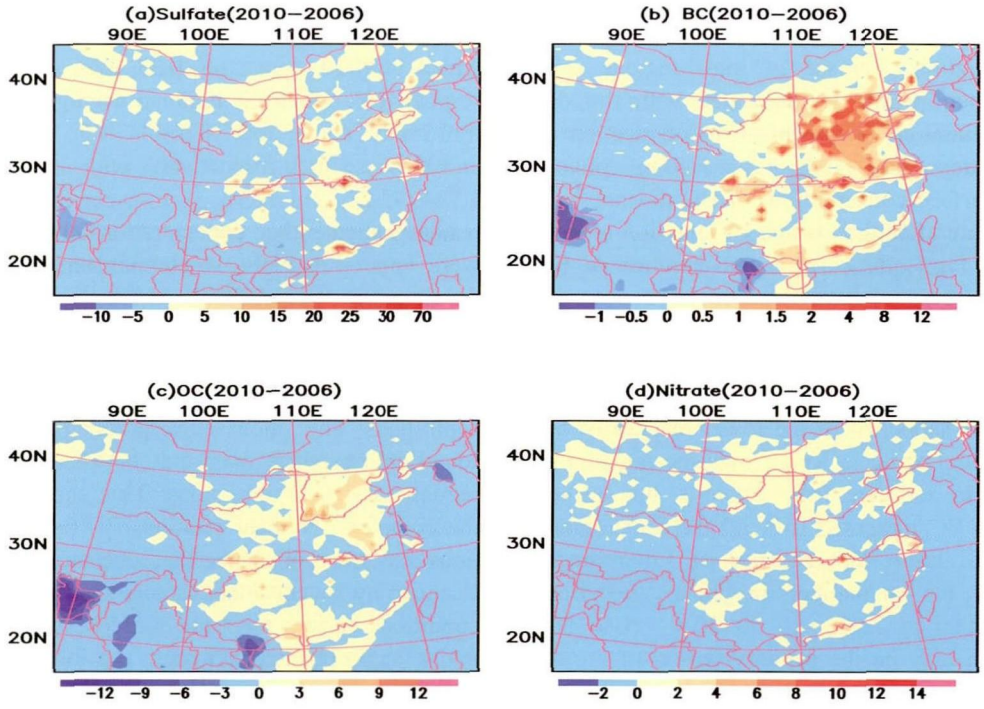

Figure 5. Distributions of the annual mean surface concentrations biases between simulations of two emission sources. (a) Sulfate; (b) BC; (c) OC; (d) Nitrate. Units: μg m^{-3}.

areas of central and eastern China, compared to that in 2006. Nonetheless, it presented significant increase in a few areas like YRD, PRD, Shandong Peninsula and Wuhan, among which the largest increase happened in Wuhan with the value of 69.28 μg m^{-3}. Induced by the large increase over the regions mentioned above, the regional average surface concentration of sulfate increased over East and North China (Table 3). We may find that the emissions of SO_2 in Wuhan in 2010 increased largely and even reached the value of 233.63 t km^{-2}, which gave rise to the big increase in surface concentration of sulfate. According to what was shown by the environment quality bulletin that SO_2 emissions of Wuhan in 2006 and 2010 were 9.28 t and 13.96 t, respectively, we

Table 3. The precursor emission, aerosol concentrations, column burdens, AOD and their change rates in different defined domains.

		East China		Central China		North China		South China	
		2010	2010–2006 2006 (%)	2010	2010–2006 2006 (%)	2010	2010–2006 2006 (%)	2010	2010–2006 2006 (%)
Average Emissions ($Gg\ year^{-1}$)	SO_2	11100.760	−16.23%	5675.960	−10.60%	3961.670	15.64%	4753.200	−13.71%
	BC	705.160	0.45%	305.910	−11.83%	243.790	−1.23%	299.970	−9.52%
	OC	1211.200	−2.24%	657.510	-6.91%	403.490	−19.87%	641.670	−16.38%
	NO_X	12263.190	49.96%	3300.080	0.47%	4579.210	53.36%	4204.560	11.71%
Average Column Burdens ($mg\ m^{-2}$)	Sulfate	4.020	−56.13%	2.440	−45.57%	1.970	−15.78%	2.270	−62.00%
	BC	0.590	57.53%	0.320	34.58%	0.210	74.72%	0.220	11.20%
	OC	2.350	6.82%	1.910	4.44%	1.520	4.52%	1.670	−3.00%
	Nitrate	1.510	−0.98%	1.320	−0.75%	1.260	−0.60%	1.280	−1.31%
Average Surface Concentrations ($mg\ m^{-3}$)	Sulfate	4.020	22.03%	1.990	7.30%	1.470	13.05%	1.410	−3.00%
	BC	1.600	102.15%	0.800	68.43%	0.560	90.72%	0.552	52.10%
	OC	2.820	13.35%	1.750	18.45%	0.960	14.64%	1.240	1.00%
	Nitrate	0.820	29.38%	0.460	13.86%	0.350	15.95%	0.330	11.30%
Aerosol Optical Depth (AOD)	Sulfate	0.027	−59.09%	0.015	−50.00%	0.012	−20.18%	0.015	−63.00%
	BC	0.030	50.00%	0.002	100.00%	0.001	60.99%	0.001	−50.00%
	OC	0.022	4.76%	0.018	0	0.014	2.78%	0.020	−17.00%
	Nitrate	0.014	−6.67%	0.013	0	0.011	−0.73%	0.016	0

Notes: (2010-2006)/2006 represents the change rate from 2006 to 2010 and it is computed by the original values without approximations.

may presume that the emission source in 2010 overestimate the SO_2 emissions in Wuhan. As is known, the surface concentration decrease in most regions over central and eastern China is associated with the decrease of SO_2 emissions arising from the conduction of energy-conservation and emission-reduction policy. However, the surface concentration of BC did not vary like that of sulfate after the change of emission source. As is shown in Fig.5, the surface concentration of BC presented an apparent upward trend in 2010 over the analysis domain with the amplitude of 0.5 $\mu g\ m^{-3}$ approximately when comparing simulation results in 2006 and 2010. Among the analysis domain, the most pronounced increase happened in the lower reaches of the Huanghe River and Yangtze River, with the maxima of 10.90 $\mu g\ m^{-3}$. In addition, from Table 3, the surface concentration of BC increased over all the four regions on average, too. From the statistics of Environment Science Institute, the car numbers in China varied from 3.088×10^7 in 2006 to 7.722×10^7 in 2010 (Bao and Zhao [28]), which may be the primary reason of the increase of BC concentration. In light of the similarities of BC and OC emissions, their surface concentration variation distributions were roughly alike. The surface concentration of OC showed a sharp increase in Sichuan Basin, together with the lower reaches of Huanghe River with the extreme of 11.52 $\mu g\ m^{-3}$. Regarding to the variations in the surface concentrations of nitrate over East China, Central China and South China, there were a few scattered regions in which the nitrate concentrations increased apparently corresponding to the distributions of NO_x emissions and the extreme appeared in Wuhan with the value of 12.50 $\mu g\ m^{-3}$. Nonetheless, the variations in nitrate concentration were mostly within the range of −2.0 to 2.0 $\mu g\ m^{-3}$ and the regional average values all showed an increasing trend.

Column burden is the vertical integral of the aerosol mass of per square meter from the surface to the top of the atmosphere. The distributions of the change in aerosol column burdens were similar to that in aerosol surface concentrations (figure omitted). By comparing the column burdens of aerosols in 2006 and 2010, we can find the same increase of different aerosols in regions such as Shanghai, Wuhan, Jiangsu, etc. This can be explained by that these regions are of developed industry, dense population and rapid economic development, which results in large quantities of emitted anthropogenic aerosols (Gao et al.[29]). From the regional average values shown in Table 3, however, we may find the changes in aerosol column burdens and surface concentrations showed some differences, especially for sulfate. This may be influenced by vertical distributions.

Table 2 shows the total emissions and annual mean concentrations averaged over the analysis domain calculated from two kinds of emission sources. It can be seen that the regional average column burdens of sulfate, BC, OC and nitrate over central and eastern China in 2010 were 2.68, 0.33, 1.86 and 1.34 $mg\ m^{-2}$, respectively, While the regional average surface concentrations of above four aerosols were 2.22, 0.87, 2.65, and 0.49 $\mu g\ m^{-3}$, respectively, with extremes of 71.61, 12.34, 12.21, and 12.96 $\mu g\ m^{-3}$, correspondingly. These results indicate that concentrations of sulfate and OC were larger relatively, in comparison with that of BC. As shown in the two kinds of emission sources and the change rate in the table, the variation trend of aerosol concentrations did not match well with that of the emissions while correlations between aerosol surface concentrations and their corresponding emissions were found to be better than that between aerosol column burdens and their emissions. This can be attributed to the complex nonlinear relationships between concentrations and emissions since the concentrations are also influenced by several other factors such as meteorological conditions besides emission sources. Among the several major anthropogenic aerosols, the increase in regional average surface concentrations of BC ranked first, followed by nitrate, sulfate and OC in turn with the change rates of 13.25%, 81.56%, 10.57% and 20.83%, respectively. Compared to those in 2006, although the annual average surface concentration of sulfate over the analysis domain in 2010 increased subtly, the column burden of sulfate in 2010 declined obviously, for the change rate of annual average column burdens in two years was −52.20%. In terms of the total emission differences between the two years over the analysis domain, BC and OC emissions in 2010 presented a minor reduction, while their average surface concentrations and column burdens increased especially for BC. The emission change of BC varied much among different regions (Fig.3), which decreased the average change value of the total area. However, the regional heterogeneity of the emission may induce large variations in surface concentration and column burden of BC. In doing so, concentrations have close relations to not only emission amounts but also emission distributions, which brings about the inconsistency between emissions and concentrations and makes the problem complicated.

Figure 6 shows the vertical distributions of aerosol concentrations over the analysis domain obtained from two different emission sources. In general, concentrations of the four kinds of aerosols decreased monotonically with altitudes except sulfate concentrations in several layers in 2006. In each layer, sulfate concentrations simulated in 2010 were mostly smaller than that in 2006, albeit the exceptions near the surface layer. This can explain why the average surface concentrations of sulfate increased in 2010 while the average column burdens of sulfate did not. Above 700 hPa, differences between the simulated results of two emission sources were minimal. At altitudes below 700 hPa, however, the BC and OC concentrations in 2010 were

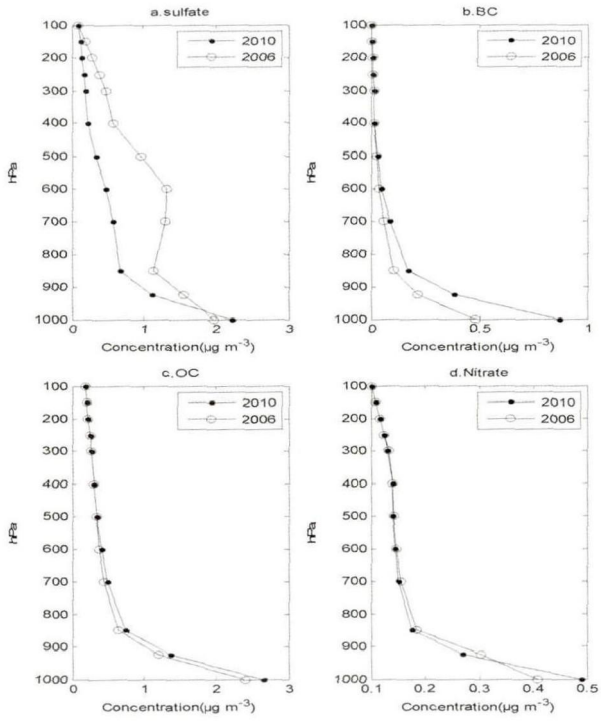

Figure 6. Distributions of vertical concentrations of aerosols over central and eastern of China. (a) Sulfate; (b) BC; (c) OC; (d) Nitrate. Units: μg m^{-3}.

larger than that in 2006 and this characteristic was more apparent for BC. Unlike other aerosols mentioned above, the vertical distributions of nitrate concentrations obtained from two simulations (in 2006 and in 2010) varied little. In addition, the nitrate concentration varied in opposite trend at the first layer and other layers, which resulted in its little variations in column burden in Table 2.

3.3 *Distributions of aerosol optical depth*

The regional average value of the mixture of the four kinds of aerosols in 2006 was 0.074, with a contribution of 0.04 from sulfate. The AOD of the mixture of the four kinds of aerosols varied from 0.03 to 0.21 in the central and eastern China with the same magnitude and similar distribution trend of the study by Huang et al. [30] in 2007 albeit some underestimates in Sichuan Basin.

Figure 7 shows the distribution of differences between aerosol optical depth (AOD) simulated by two emission sources. The AOD distributions are in good agreement with the column burdens of aerosols. The AOD of sulfate in 2010 broadly presented a downward trend in comparison with that in 2006. From Table 3, the average decrease in East China, Central China, North China and South China were 59.09%, 50.0%, 20.18% and 63.00%, respectively. As is shown in the figure, the decrease range was between 0.04 and 0.08 over East China as well as regions adjacent to Sichuan Basin and the reduction even reached 0.12 in some regions. The AOD of BC showed a striking increase over North China and East China with the maximum value

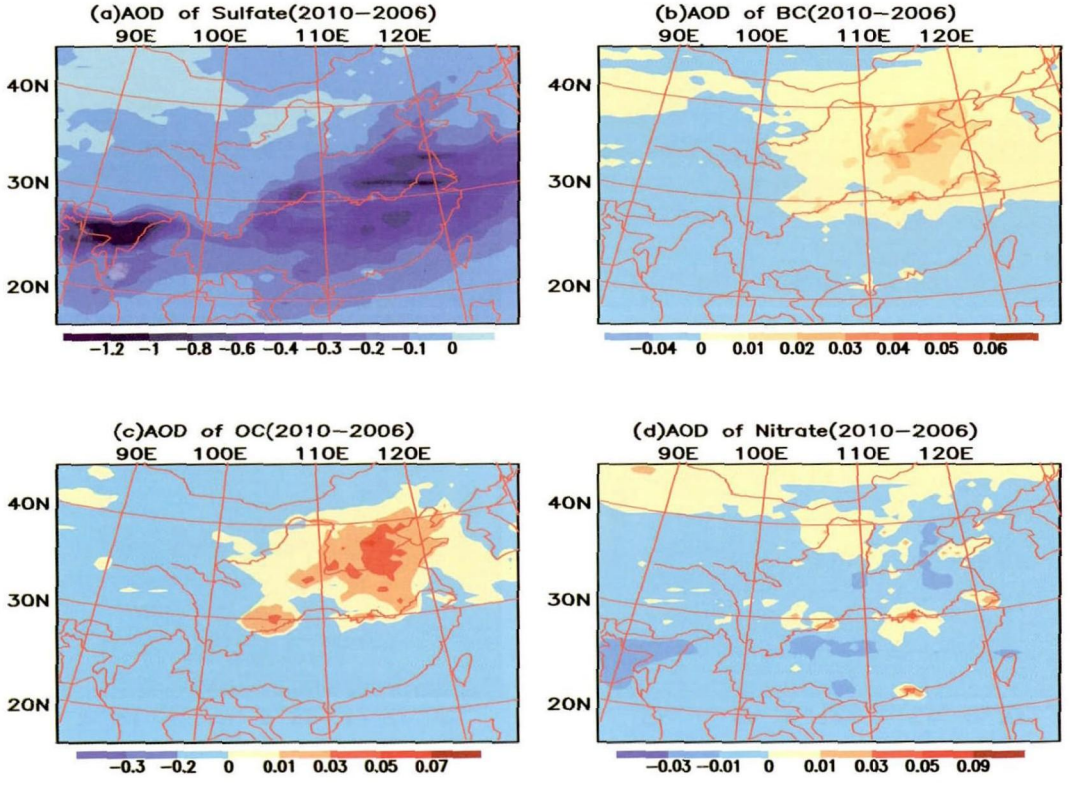

Figure 7. Distributions of the AOD biases between simulations of two emission sources (10^{-1}). (a) Sulfate; (b) BC; (c) OC; (d) Nitrate.

of 0.006 and the average increase of 60.99% and 50.00%, respectively. Also evident was the increase in PRD while it was quite different over a few other regions on the southeast coast of China where the AOD of BC declined. Although changes in the AOD of OC were similar to that of BC in distribution, the former had larger amplitude of increase with the maximum value of 0.007. Regions in which the AOD of nitrate increased in high values were still separated over central and eastern China and the biggest value reached 0.008. In spite of this, the AOD of nitrate varied little over most regions within the range of −0.001 to 0.001 and its average changes in different domains were small too (Table 3).

In summary, the AOD of BC and OC increased over most regions of East China and North China, particularly the regions with high degree of industrialization, fast growth of economic development and high population density. While for the AOD of sulfate, it broadly decreased or varied little. From the statistic results of regional average over central and eastern China, we may find that the AOD of BC increased, the AOD of OC and nitrate decreased slightly and the AOD of sulfate decreased clearly (Table 3).

3.4 *Radiative forcings of aerosols and their effects on surface air temperature*

The all-sky radiative forcing (all-sky RF, equals to the RF later) of all the four kinds of anthropogenic aerosols together and that of BC alone at the TOA are shown in Fig.8. Their respective influences on the surface air temperature are displayed too. Obviously, the RF of BC at the TOA was positive with the extreme of 1.12 W m^{-2}, which resulted from its strong absorption of incoming short wave radiation. Also evident is the warming effect of BC over many areas; with the maximum value of 0.20 K. Differently, sulfate, nitrate and OC played an important role in scattering solar radiation, which gave rise to their negative RF at the TOA. The negative RF of scattering aerosols outweighed the positive RF of absorption aerosols, resulting in the net negative RF of these four anthropogenic aerosols. Consistent to this, when all the aerosols are considered, the net effect of cooling over most regions in the analysis domain was also demonstrated in the figure. Hence, the viewpoint that BC is the second significant factor contributing to climate warming proposed in the study (Bai and Wang[31]) is incomplete, since it is obtained by considering the climate effect of BC only, rather than all the anthropogenic aerosols including scattering aerosols. In fact, not only the concentration of BC but also those of other aerosols have been increasing in recent decades and to our knowledge, there have not been any study pointing out that the warming caused by BC outweighs the cooling due to scattering aerosols globally or regionally. It is worth noticing that until now, all aerosols together have been considered to exert a cooling effect all the time, yet the climate effects have varied with the change of source emissions.

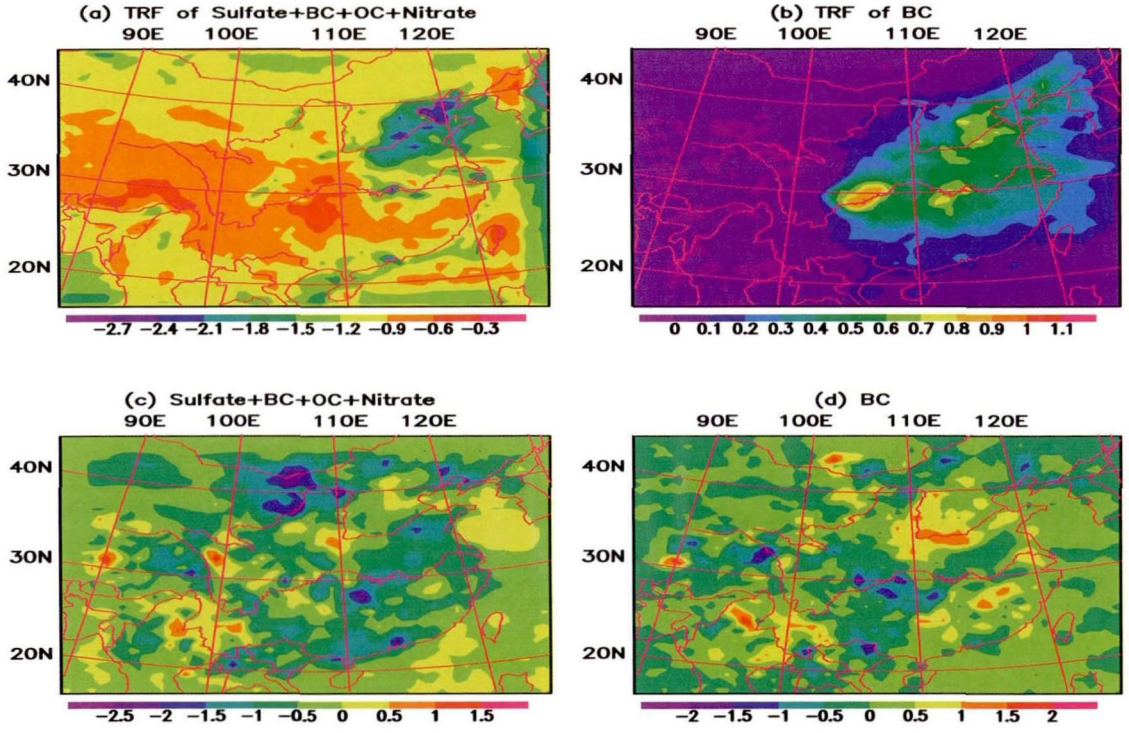

Figure 8. The radiative forcings of aerosols at the TOA (Units : W m^{-2}) and the surface air temperature response to aerosols in 2010 (Units: 10^{-1} K)(a) TRF: Sulfate+BC+OC+Nitrate; (b) TRF: BC; (c) dT: Sulfate+BC+OC+Nitrate; (d) dT: BC.

Figure 9 shows the change in the annual mean RF of aerosols at the TOA in simulations of 2010 as compared to that of 2006. With the emission source changed, the RF of sulfate weakened generally except a few regions over North China, which was in agreement with the distributions of the change in the AOD of sulfate. In spite of this, among the four aerosols mentioned before, the absolute value of the RF of sulfate still came first. In terms of the change in the RF of BC, the increase occurred in most regions over East and North China in a scope of 0 to 0.5 W m^{-2} and relatively large increases were in the areas between Yangtze and Huanghe Rivers with the maxima of 0.6 W m^{-2}. Over many areas of East China and Central China, meanwhile, the RF of OC mainly strengthened from −0.4 to 0 W m^{-2}. There was an area in which the RF of OC was enhanced largely in an interval of −0.4 to −0.8 W m^{-2} between the lower reaches of Huanghe River and Sichuan Basin. In contrast, the RF of OC was suppressed over the regions in the southeast coastal part of Chinese mainland. Large uncertainty existed concerning the variations in the RF of nitrate. However, the change in the RF of nitrate was subtle with the range from −0.2 to 0.2 W m^{-2}. We can see from the figure that the RF of nitrate generally weakened over most regions of the analysis domain and the largest variation occurred in regions between Yangtze and Huanghe River.

The changes in the column radiative forcing (CRF)

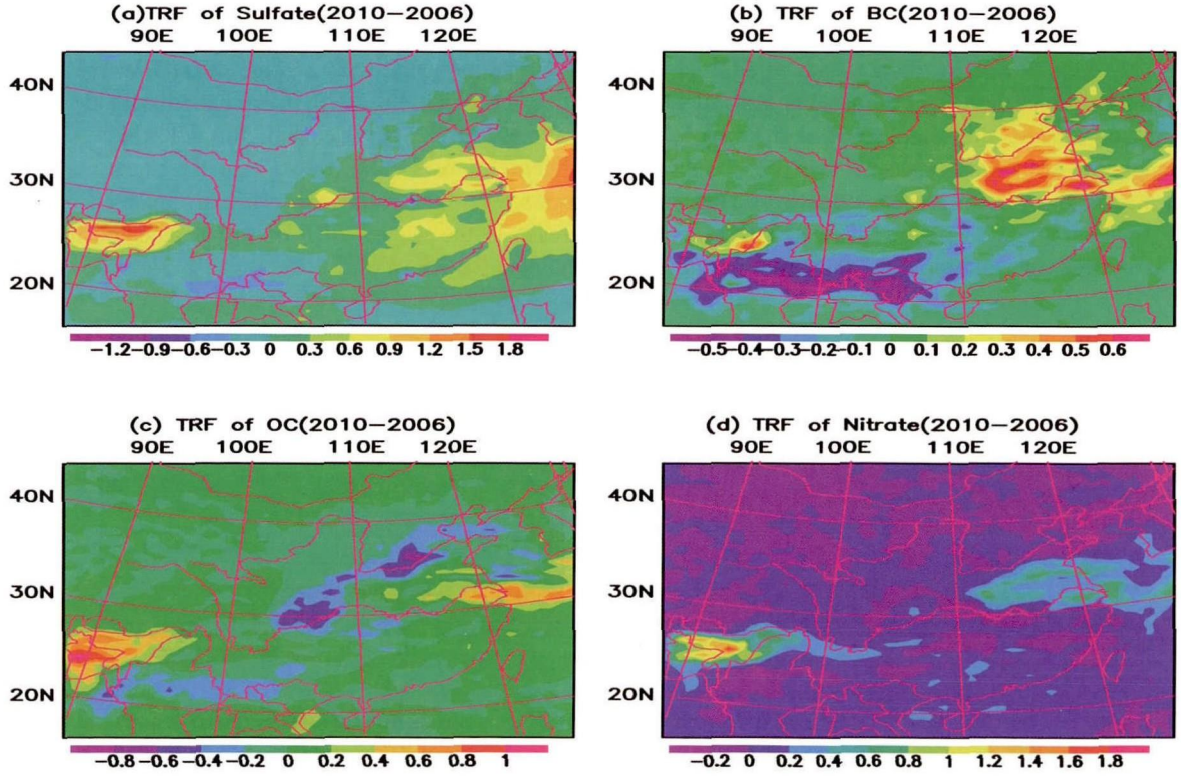

Figure 9. Distributions of the radiative forcings biases at the top of the atmosphere between the simulations of two emission sources. (a) Sulfate; (b) BC; (c) OC; (d) Nitrate. Units: W m^{-2}.

and surface radiative forcing (SRF) of BC in 2010 as compared to those in 2006 are shown in Fig.10, respectively. The CRF of BC strengthened over most regions of central and eastern China, especially the middle and lower reaches of Huanghe River, and so did the SRF of BC. It was manifested that the CRF of BC was positive, the SRF of BC was negative and the increased value of its CRF outweighed its increase in SRF in many areas, which made the net RF of BC enhanced in most areas and suppressed in a few areas. By comparing Fig.10a with Fig.10c, we may find that the distribution of the changes in the CRF of the four kinds of aerosols together matched well with that of BC alone even with the same magnitude. According to this characteristic, it can be concluded that BC contributes the largest to the CRF of all the aerosols together. From Fig.10d which shows the distribution of differences between the net RF of all kinds of aerosols in 2010 and that in 2006, several features are concluded as described below. The net negative RF at the TOA of all the four kinds of anthropogenic aerosols together in 2010 weakened over most regions of South China and East China and the range of weakening was from 0.50 to 2.0 W m^{-2}. Conversely, over some regions in North China, meanwhile, the net negative RF of all aerosols strengthened with the extreme of −0.70 W m^{-2}. It was sulfate which was the

most significant contributor to the RF of all aerosols together that caused the decrease in the net RF value of all the four kinds of aerosols over South and East China. So did that over North China.

Generally speaking, the distributions of the change in the AOD of aerosols are in consistence with that in the RF of aerosols in most regions where the increase in the AOD of aerosols were accompanied by the increase in the RF of aerosols. Nonetheless, the variation values of the RF were not proportional to that of the AOD and even some opposite variations between them occurred in a few regions. Table 4 shows the annual mean AOD and RF of the aerosols as well as their change rates all averaged over the analysis domain both in 2006 and 2010. From the change rates of the AOD and RF , we can find that the AOD of aerosols present a nonlinear correlation with their all-sky RF as well as their clear-day RF. This is due to the fact that in addition to the AOD of aerosols, the RF of aerosols is also affected by the surface albedo, the temperature and pressure of the atmosphere (Mao and Li [32]; Li et al.[33]). As is demonstrated in the table, the clear-day RF of each kind of aerosol was stronger than its all-sky RF. The annual mean all-sky RF of sulfate, BC, OC and nitrate in 2010, averaged over the analysis region were −0.64, 0.29, −0.41 and −0.33 W m^{-2}, respectively while in 2006, the values came to be −0.78, 0.26, −0.35 and −0.40 W m^{-2}. When all the four kinds of anthropogenic aerosols were considered, the all-sky RF was −1.1 W m^{-2} in 2010 and −1.65 W m^{-2} in 2006. Furthermore, simply adding the all-sky RF of the four aerosols in 2006 can reach the value of −1.27 W m^{-2}, which was far more different from the value of −1.65 W m^{-2} calculated in case01 with all kinds of aerosols considered. Consequently, the climate effect of a variety of aerosols is not simply equal to the addition of the climate effect of each kind of aerosol. In addition, compared to that in 2006, the varied values in the RF of the four aerosols in 2010 are computed as follows. The negative RF of sulfate and nitrate in 2010 weakened with the value of 0.14 W m^{-2} and 0.07 W m^{-2}, respectively; the positive RF of BC in 2010 strengthened with a value of 0.03 W m^{-2}; the negative RF of OC in 2010 strengthened with the value of 0.06 W m^{-2}. When considering all kinds of anthropogenic aerosols, the decrease of the negative RF came to 0.55 W m^{-2}, which was primarily induced by the decrease in the negative RF of sulfate and the increase in the positive RF of BC.

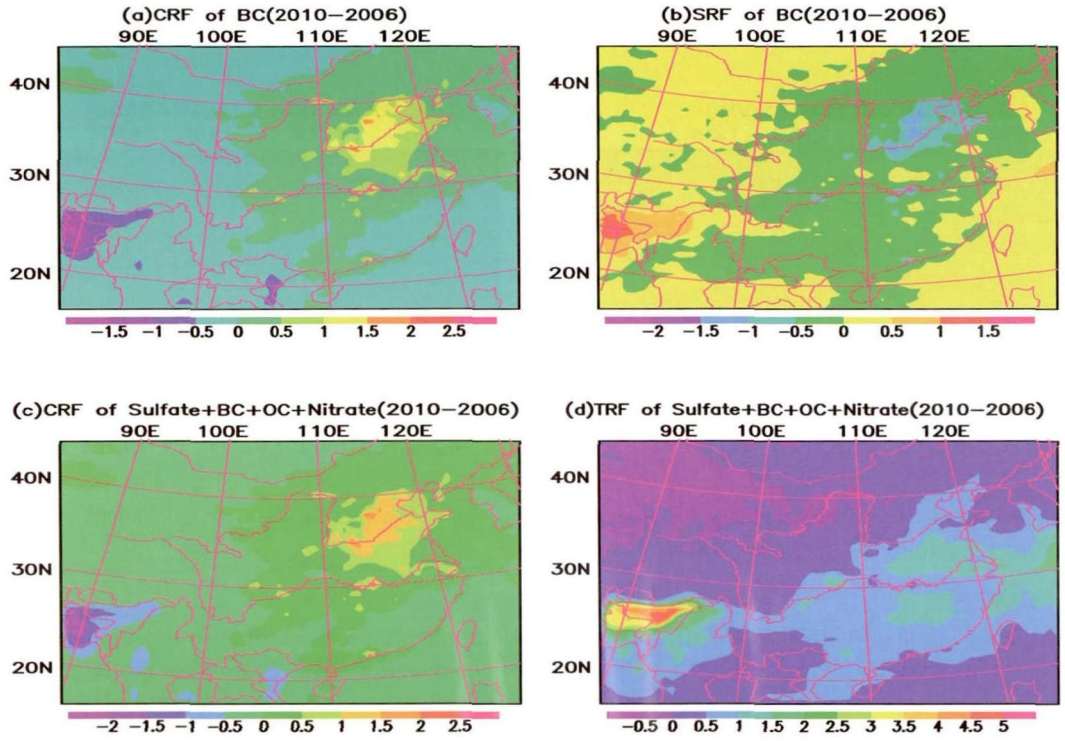

Figure 10. The CRF and SRF biases of aerosols between the simulations of two emission sources. (a) CRF of BC; (b) SRF of BC; (c) CRF of Sulfate+BC+OC+Nitrate; (d) SRF of Sulfate+BC+OC+Nitrate. Units: W m^{-2}.

Figure 11 shows the differences between the effect on the surface air temperature of aerosols simulated in 2010 and that in 2006. Compared to that in 2006, the cooling due to sulfate in 2010 was enhanced over several areas near Sichuan Basin, North China, as well as a few regions of South China, while it weakened over most areas of North China, Central China and East China. This was in good agreement with the changes of SO_2 emissions over these areas. The weakened cooling effect of sulfate produced a warmer surface air temperature over central and eastern China and the largest increase value come to 0.25 K. As for BC, over most of

the areas of China, its warming effect enhanced with the largest amplification more than 0.2 K. Near Sichuan Basin, however, as well as the regions of South China, the warming due to BC in 2010 was weaker, which agreed to the decreasing trend in the RF of BC in 2010 over these areas. Additionally, the cooling effect of OC enhanced in some areas of East China, obviously in the regions between Yangtze and Huanghe Rivers. Roughly speaking, the effect on the surface air temperature caused by nitrate varied little after the changing in emission sources within the range of 0.1 K (figure omitted). Only over the regions of Central China did the

Table 4. Regional average values of the aerosol optical depth and radiative forcings of the anthropogenic aerosols over central and eastern China.

Aerosols	Year	AOD	All-sky RF (W m^{-2})	Clear-sky RF (W m^{-2})
Sulfate	2010	0.017 (0.075)	−0.64 (−2.49)	−0.93 (−3.65)
	2006	0.039 (0.144)	−0.78 (−2.01)	−1.30 (−3.43)
	(2010–2006)/2006	−55.40%	−18.33%	−28.24%
BC	2010	0.002 (0.008)	0.29 (1.12)	0.29 (1.44)
	2006	0.001 (0.004)	0.26 (0.95)	0.24 (0.86)
	(2010–2006)/2006	14.25%	12.59%	22.02%
OC	2010	0.019 (0.033)	−0.41 (−0.98)	−0.65 (−1.19)
	2006	0.019 (0.031)	−0.35 (−1.17)	−0.61 (−1.98)
	(2010–2006)/2006	−3.88%	15.55%	5.99%
Nitrate	2010	0.014 (0.022)	−0.33 (−0.63)	−0.46 (−0.95)
	2006	0.014 (0.018)	−0.40 (−1.08)	−0.57 (−1.64)
	(2010–2006)/2006	−1.99%	−16.33%	−19.99%
Sulfate+BC+OC+Nitrate	2010	0.051 (0.133)	−1.10 (−2.81)	−1.79 (−4.17)
	2006	0.074 (0.181)	−1.65 (−3.51)	−2.74 (−5.32)
	(2010–2006)/2006	−30.32%	−33.08%	−34.72%

Notes: The values in brackets are the maximum values of AOD and the extreme values of radiative forcing of different aerosols (the maximum value for the radiative forcing of BC and the minimum values for that of other aerosols); (2010–2006)/2006 represents the change rate from 2006 to 2010 and it is computed by the original values without approximation.

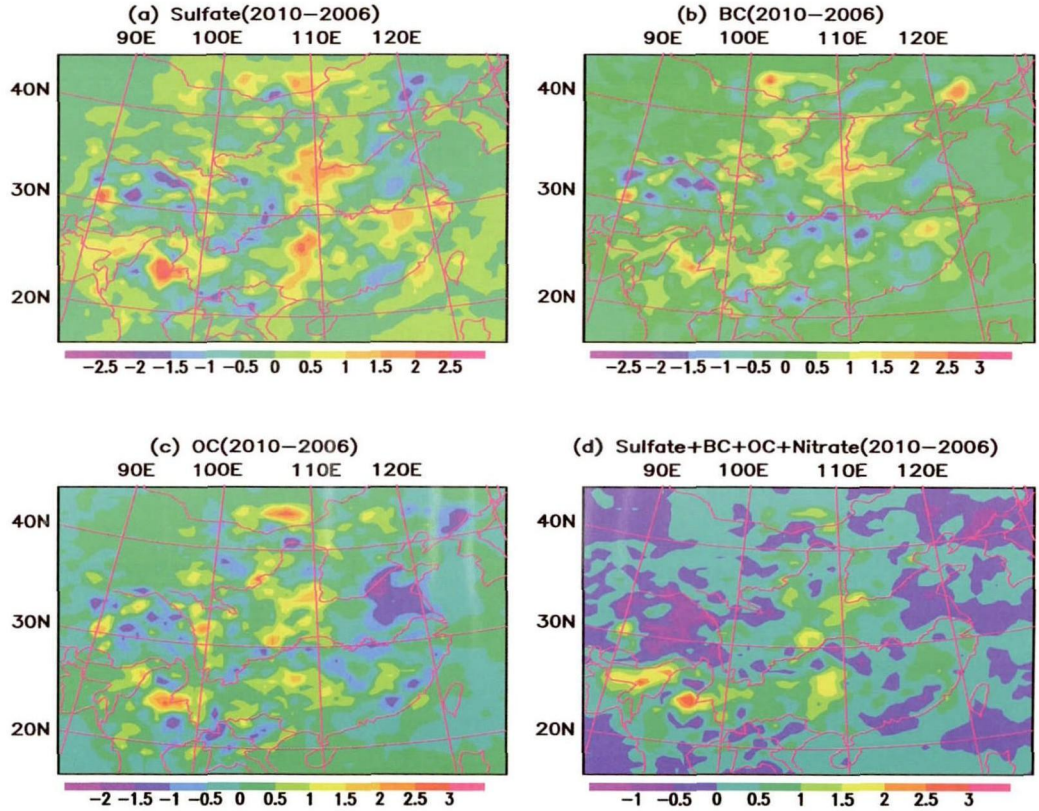

Figure 11. Distributions of the surface air temperature variation biases between the simulations of two emission sources. (a) Sulfate; (b) BC; (c) OC; (d) Sulfate+BC+OC+Nitrate. Units: 10^{-1} K.

cooling due to nitrate occurred, which meant that over Central China, nitrate produced "an effect on increasing the surface air temperature" with the emission sources changed and the largest increase value being 0.36 K. The cooling caused by nitrate in 2010 over some regions of East China and North China was still enhanced

In brief, after changing the emission source, the variations in the surface air temperature induced by the four aerosols were generally in agreement with their RF variations over most areas. However, in some regions on the east coast of China, the net RF of all the four aerosols weakened while the cooling caused by all the aerosols strengthened instead. In light of this, the relationship between temperature changes responding to aerosols and the RF of aerosols was nonlinear and besides RF, the temperature changes were also affected by factors such as precipitation and atmosphere circulation.

Since the influences of the aerosols on the surface air temperature varied regionally, we have discussed the temperature change caused by anthropogenic aerosols in different regions. Table 5 shows the regional average values of the temperature change caused by different aerosols in the four regions. Obviously, in terms of averaged values, sulfate and OC induced cooling over all of the four regions. The regional average temperature changes caused by sulfate and OC over the whole central and eastern China were −0.036 K and −0.026 K, respectively and the extreme values were −0.26 K and −0.27 K, respectively. Over East China, meanwhile, BC tended to increase the temperature and the regional average increase in 2010 due to BC was 0.027 K. When it came to nitrate, the uncertainties in regional changes were large. When all the four anthropogenic aerosols were considered, it tended to decrease the surface air temperature over all of the four regions and the regional average reductions were −0.064, −0.053, −0.053 and −0.059 K over East China, Central China, North China and South China, respectively. Over central and eastern China, the average decrease in temperature caused by all the aerosols in 2010 was −0.096 K, together with the extreme of −0.285 K while in 2006 those were −0.063 K and −0.256 K. From these values, we can learn that the changes in emission sources weakened the cooling effect of aerosols on average. Here are two reasons. On one hand, the warming due to BC was strengthened. On the other, the cooling caused by the scattering aerosols was weakened. In some sense, the net cooling effect of all the anthropogenic aerosols can outweigh the warming effect due to greenhouse gases and its weakening may bring in an intense increase of the temperature in future. In order to improve the air quality, the Chinese government has put forward the "emission-reduction" policy, which has led to the decrease in the concentrations of SO_2 and PM_{10}. But what has to be mentioned is that the reduction of "carbon emissions" (including CO_2 and BC) must be considered at the same time to avoid the enhancement of greenhouse effect accompanied with the reduction of air pollution.

Table 5. The average air temperature variations caused by anthropogenic aerosols in different areas.

Aerosol	Year	Surface air temperature change over different regions				
		East China	Central China	North China	South China	Central and eastern China
Sulfate	2010	−0.037	−0.020	−0.050	−0.039	−0.036
		(−0.144)	(−0.222)	(−0.264)	(−0.179)	(−0.264)
	2006	−0.033	−0.021	−0.045	−0.046	−0.036
		(−0.169)	(−0.206)	(−0.228)	(−0.186)	(−0.228)
BC	2010	0.027	−0.002	0.006	0.007	0.010
		(0.149)	(0.136)	(0.130)	(0.203)	(0.203)
	2006	0.018	−0.010	−0.029	−0.002	−0.005
		(0.140)	(0.106)	(0.115)	(0.142)	(0.142)
OC	2010	−0.041	−0.022	−0.032	−0.0126	−0.026
		(−0.145)	(−0.268)	(−0.187)	(−0.145)	(−0.268)
	2006	−0.0008	−0.050	−0.050	−0.029	−0.032
		(−0.191)	(−0.300)	(−0.176)	(−0.221)	(−0.300)
Nitrate	2010	−0.002	0.009	−0.019	−0.007	−0.004
		(−0.108)	(−0.202)	(−0.173)	(−0.202)	(−0.202)
	2006	0.005	−0.035	−0.035	−0.044	−0.029
		(−0.170)	(−0.236)	(−0.186)	(−0.251)	(−0.251)
Sulfate+BC+OC+Nitrate	2010	−0.064	−0.053	−0.053	−0.059	−0.063
		(−0.221)	(−0.221)	(−0.256)	(−0.206)	(−0.256)
	2006	−0.083	−0.098	−0.098	−0.102	−0.096
		(−0.261)	(−0.285)	(−0.285)	(−0.278)	(−0.285)

Notes: The values in brackets are the extreme values of aerosols changing the regional air temperature (the maximum value for BC and the minimum values for other aerosols.

4 CONCLUSIONS AND DISCUSSIONS

In this paper, we analyzed the distributions and radiative effects of anthropogenic aerosols and their sensitivities to emission sources using the RIEMS2.0 model, forced by the emission inventory in 2006 and the high-resolution emission inventory in 2010. The main conclusions drawn are as follows:

(1) When using the emission source in 2010 instead of that in 2006, the surface concentrations of sulfate generally presented a downward trend with a scope of −5 to 0 gm^{-3} over central and eastern China. Over many areas of the analysis domain, the surface concentrations of both BC and OC increased obviously in a range of 0 to 0.5 gm^{-3} and 0 to 3.0 gm^{-3}, respectively. The extreme value of the increase in the surface concentration of BC reached 10.9 gm^{-3}. When it came to nitrate, its surface concentration varied in most areas with a range of −2.0 to 2.0 gm^{-3}.

(2) Compared with that in 2006, the AOD of sulfate in 2010 substantially decreased in a range of 0.04 to 0.08 near Sichuan Basin and over East China, even reaching 0.12 in some regions. The AOD of BC and OC revealed an increase over East China and North China and the extreme values of the increase were 0.006 and 0.007, respectively.

(3) The annual average all-sky RF of sulfate, BC, OC and nitrate over central and eastern China in 2010 were −0.64, 0.29, −0.41 and −0.33 W m^{-2}, respectively, while those in 2006 were −0.78, 0.26, −0.35 and −0.40 W m^{-2} respectively. When considering all the anthropogenic aerosols together, the net all-sky RF were −1.1 W m^{-2} in 2010 and −1.65 W m^{-2} in 2006. The RF of BC and OC in 2010 strengthened over many areas of East China compared with that in 2006. The net negative RF of the four anthropogenic aerosols in 2010 was weaker than that in 2006 over South China and East China.

(4) After substituting the emission source in 2006 with that in 2010, the climate effects of aerosols changed as well and the distributions of changes in the surface air temperature variations induced by different aerosols varied widely. The decrease in sulfate brought about an increase in the temperature over central and eastern China with a maximum of 0.25 K. Additionally, the increase of BC over most of the areas enhanced its warming. However, the increase of OC strengthened its cooling effect over similar areas, particularly the areas between the Yangtze and Huanghe River, which offsets the effect caused by the increase of BC to a certain extent. The changes in the surface air temperature caused by nitrate varied between different domains and its influence was relatively small. The changes in the surface air temperature caused by nitrate were similar to that by sulfate mentioned above, albeit the respectively minor value. The annual average surface air temperature change caused by the four aerosols together was −0.096 K in 2006 and −0.063 K in 2010, accompanied with the largest variation of −0.285 K and −0.256 K, respectively. On the whole, the changes of emission sources lowered the cooling effect of anthropogenic aerosols, which can be explained by the fact that the warming effect of BC has been enhanced while the cooling effect of scattering aerosols has been weakened.

We mainly considered the external mixture state of aerosols in this paper and the results may be different in some way when using other mixture states.

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