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Direct and semi-direct radiative effects of anthropogenic aerosols in the Western United States: Seasonal and geographical variations according to regional climate characteristics

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Abstract The direct and semi-direct radiative effects of anthropogenic aerosols on the radiative transfer and cloud fields in the Western United States (WUS) according to seasonal aerosol optical depth (AOD) and regional climate are examined using a regional climate model (RCM) in conjunction with the aerosol fields from a GEOS-Chem chemical-transport model (CTM) simulation. The two radiative effects cannot be separated within the experimental design in this study, thus the combined direct- and semi-direct effects are called radiative effects hereafter. The CTM shows that the AOD associated with the anthropogenic aerosols is chiefly due to sulfates with minor contributions from black carbon (BC) and that the AOD of the anthropogenic aerosol varies according to local emissions and the seasonal low-level winds. The RCM-simulated anthropogenic aerosol radiative effects vary according to the characteristics of regional climate, in addition to the AOD. The effects on the top of the atmosphere (TOA) outgoing shortwave radiation (OSRT) range from -0.2 Wm^{-2} to -1 Wm^{-2}. In Northwestern US (NWUS), the maximum and minimum impact of anthropogenic aerosols on OSRT occurs in summer and winter, respectively, following the seasonal AOD. In Arizona-New Mexico (AZNM), the effect of anthropogenic sulfates on OSRT shows a bimodal distribution with winter/summer minima and spring/fall maxima, while the effect of anthropogenic BC shows a single peak in summer. The anthropogenic aerosols affect surface insolation range from -0.6 Wm^{-2} to -2.4 Wm^{-2}, with similar variations found for the effects on OSRT except that the radiative effects of anthropogenic BC over AZNM show a bimodal distribution with spring/fall maxima and summer/winter minima. The radiative effects of anthropogenic sulfates on

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TOA outgoing longwave radiation (OLR) and the surface downward longwave radiation (DLRS) are notable only in summer and are characterized by strong geographical contrasts; the summer OLR in NWUS (AZNM) is reduced (enhanced) by 0.52 Wm^{-2} (1.14 Wm^{-2}). The anthropogenic sulfates enhance (reduce) summer DLRS by 0.2 Wm^{-2} (0.65 Wm^{-2}) in NWUS (AZNM). The anthropogenic BC affect DLRS noticeably only in AZNM during summer. The anthropogenic aerosols affect the cloud water path (CWP) and the radiative transfer noticeably only in summer when convective clouds are dominant. Primarily shortwave-reflecting anthropogenic sulfates decrease and increase CWP in AZNM and NWUS, respectively, however, the shortwave-absorbing anthropogenic BC reduces CWP in both regions. Due to strong feedback via convective clouds, the radiative effects of anthropogenic aerosols on the summer radiation field are more closely correlated with the changes in CWP than the AOD. The radiative effect of the total anthropogenic aerosols is dominated by the anthropogenic sulfates that contribute more than 80% of the total AOD associated with the anthropogenic aerosols.

1 Introduction

The impact of anthropogenic aerosols on regional climate has become a topic of intense research (Boucher and Anderson 1995; Mitchell et al. 1995; Pan et al. 1997; Giorgi et al. 2002; Gu et al. 2006; Kim et al. 2006; Huang et al. 2007). However, its regional effects in the western United States (WUS) remain largely uncertain. Analyses of direct insolation in four southern China cities revealed that the increase in aerosol concentration due to increased local emissions have reduced the present-day direct insolation by over 20% of that during the period 1960–1980 (Luo et al. 2000). In an analysis of summer climate change, air pollution, and clear sky insolation over China, Xu (2001) found that increases in sulfate aerosols and the associated changes in albedo may play an important role in the summer climate pattern in eastern China characterized by *North-drought/South-flooding*. In a GCM study, Menon et al. (2002) showed that the radiative heating by absorbing aerosols such as black carbon (BC) may affect the regional atmospheric circulation and water cycle in China and India. While studies have been focused on some heavily polluted regions such as China, few studies have been focused on the western United States (WUS). Gueymard et al. (2000) reported that dusts originating from China affect the solar radiation in WUS and may alter the regional atmospheric circulation and water cycle. Thus, accurate calculation of the effects of anthropogenic aerosols on the radiative transfer and regional hydrologic cycle, especially via clouds, is crucial for global and regional climate simulations, especially for reducing uncertainties in projecting future climate change and its impacts on human society and environments.

The aerosol radiative effects on climate are complicated due to variations in aerosol types and concentrations, the feedback through other components of the climate system such as clouds, and the characteristics of regional climate (Giorgi et al. 2002; Gu et al. 2006; Kim et al. 2006). Aerosol optical properties vary according to aerosol types, and the net shortwave radiative forcing on the atmospheric column is determined by the differences between two opposite effects of reflecting (e.g., sulfates, small dust particles) and absorbing (e.g., BC, large dusts) aerosols. Previous studies of aerosol radiative effects often suffer from significant uncertainties because only a limited number of aerosol types and/or characteristics were considered. In a series of coupled regional climate-chemistry modeling, Giorgi et al. (2002) and Huang et al. (2007) found that the direct radiative forcing by anthropogenic sulfate varies according to local emissions and season. In a regional modeling study, Kim et al. (2006) showed that the direct aerosol forcing on surface

insolation and snowmelt in the Sierra Nevada depends not only on AOD but also on regional climate characteristics, especially the low-level air temperature and the frequency of cloud formation.

Due to significant orography and seasonal and geographical contrasts in climate, the impact of aerosols can vary significantly according to season and geography in the WUS. Investigations of the aerosol effects on the climate in the region has been extremely limited despite the fact that seasonal variations in the regional climate and water cycle are strongly affected by aerosol radiative forcing (e.g., Kim et al. 2006). This study attempts to investigate the direct and semi-direct effects of anthropogenic aerosols on the regional climate in WUS according to the geographical and seasonal variations in regional climate characteristics and AOD as well as in the presence of the feedback through clouds by implementing a comprehensive aerosol field and its optical properties calculated in a state-of-the-art chemistry transport model (CTM) into a regional climate model (RCM) simulations.

In the following, we present the direct and semi-direct radiative effects of anthropogenic aerosols, with further focus on scattering sulfates and absorbing BC aerosols, on the radiation and cloud fields in an RCM study in which the monthly-mean aerosol field for 2001 simulated in the GEOS-Chem CTM is prescribed. In Section 2, we describe the model structure and experimental design for this study. Sections 3 and 4 present the anthropogenic AOD field in the GEOS-Chem simulation and the impact of anthropogenic aerosols on the simulated radiation field. Section 5 presents the relationship between the overall direct aerosol radiative effect and the feedback from cloud fields, followed by conclusions in Section 6.

2 Model description and experiment design

2.1 Model description

The RCM used in this study is composed of the Mesoscale Atmospheric Simulation (MAS) model (Soong and Kim 1996; Kim 2005; Kim et al. 2006) interactively coupled with the NCEP-Oregon State University-Air Force-Office of Hydrology (NOAH) land surface scheme (Kim and Ek 1995; Chang et al. 1999). The MAS model is a primitive-equation, limited-area atmospheric model written on the σ-coordinates. Advection equation is solved using the 3rd-order accurate finite difference scheme (Takacs 1985). A modified version of the bulk microphysics scheme of Cho et al. (1989) that includes five types of hydrometeors, and the Simplified Arakawa-Schubert scheme (Pan and Wu 1995; Hong and Pan 1998) are used to compute grid-scale and convective precipitation, respectively. The effects of vertical turbulent mixing are computed using the bulk aerodynamic scheme of Deardorff (1978) at the surface, and the K-theory method within the model atmosphere. The eddy diffusivities for the K-theory method are computed using the local scheme of Louis et al. (1982) in conjunction with the asymptotic mixing length obtained in the observational study of Kim (1990) and Kim and Mahrt (1992). The 4-layer NOAH scheme coupled with the MAS model computes the land-surface processes and surface fluxes. The NOAH model predicts the volumetric soil moisture content, both frozen and unfrozen, and soil temperature within model soil layers. It also predicts the canopy-water content and snow-water equivalence at the surface. The temperature and specific humidity for calculating surface sensible and latent heat fluxes, outgoing longwave radiation, and ground heat fluxes are calculated by iteratively solving a nonlinear form of the surface energy balance equation. The MAS has been used in a number of regional climate modeling studies for the western- and continental

United States as well as for East Asia (e.g., Soong and Kim 1996; Kim 1997, 2001, 2005; Kim et al. 2002, 2006; Kim and Lee 2003). For more details of the MAS and NOAH models, readers are referred to Mahrt and Pan (1984), Pan and Mahrt (1987), Kim and Ek (1995), and Soong and Kim (1996).

Radiative transfer within the model atmosphere, including the impact of clouds and multiple types of atmospheric aerosols, is computed using the δ-2/4-stream Fu-Liou-Gu scheme modified on the basis of original Fu-Liou scheme (Fu and Liou 1992, 1993; Gu et al. 2003, 2006). This radiation scheme has been recently incorporated in MAS and has been successfully employed in regional climate studies associated with aerosols (Kim et al. 2006). The scheme uses a combination of the δ-4-stream approximation for the solar flux (Liou et al. 1988) and the δ-2/4-stream approximation for the infrared flux to achieve a balance between accuracy and computational efficiency (Fu et al. 1997). The Fu-Liou-Gu scheme includes the formulation for calculating the direct radiative effects of 18 types of aerosol and optical properties of liquid- and ice cloud particles. The aerosol types in the Fu-Liou scheme are; maritime, continental, urban, mineral dust aerosols in five size classes, insoluble, water soluble, soot, mineral dusts in four types, seas salts in two modes, and sulfates. More details on the aerosol types are presented in Gu et al. (2006). The radiative properties of aerosols, including the extinction coefficient, single-scattering albedo and asymmetry factor, are determined by their composition, shape and size distribution using the Optical Properties of Aerosols and Clouds (OPAC) database (d'Almeida et al. 1991; Tegen and Fung 1995; Tegen and Lacis 1996; Hess et al. 1998). The single-scattering properties of 18 aerosol types at 60 wavelengths within the spectral region between 0.3 μm and 40 μm are interpolated into the Fu-Liou-Gu spectral bands. Calculations of the cloud radiative forcing follow the procedure by Fu and Liou (1993) for the parameterization of the single-scattering properties of cloud particles. This scheme calculates the spectral extinction coefficient, the single-scattering albedo, and the asymmetry factor, in terms of the cloud water content, separately for ice and liquid, and the effective particle size. Calculations of the single-scattering properties of clouds require information about the particle shape and size distributions, and the indices of refraction as a function of wavelength.

The aerosol data used in this study have been simulated using the GEOS-Chem chemical transport model (CTM) version 7.02 (http://www-as.harvard.edu/chemistry/trop/geos) of coupled aerosol-oxidant chemistry. Evaluation of the GEOS-Chem aerosol data used in this study has been extensively conducted over the United States and has been presented in Park et al. (2003, 2004, 2006). Aerosols in the model include sulfate, nitrate, ammonium, carbonaceous aerosols, soil dust, and sea salt. It uses assimilated meteorological data in the NASA Goddard Earth Observing System (GEOS-3) including winds, convective mass fluxes, mixed layer depths, temperature, clouds, precipitation, and surface properties with 6-hour resolutions (3-hour for surface variables and mixing depths), $1°\times1°$ horizontal resolution, and 48 σ layers. For computational efficiency, horizontal resolutions for global CTM simulations are typically made at $2°\times2.5°$ or $4°\times5°$. Finer resolution CTM data can be obtained using the self-nesting capability of the GEOS-Chem that allows us to generate fine-resolution aerosol data from its own coarse-resolution global data using the same chemistry formulations over selected regions. This one-way nesting capability has been successfully applied to an ozone-NO_x-hydrocarbon chemistry simulation over East Asia (Wang et al. 2004) and to a coupled oxidant-PM simulation over North America (Park et al. 2006). For more details on the GEOS-Chem aerosol simulations, readers are referred to the previous publications Park et al. (2004, 2006), Alexander et al. (2005), and Fairlie et al. (2007).

2.2 Experimental design

In order to examine the direct radiative effect of anthropogenic aerosols on the radiation field in WUS, two aerosol datasets corresponding to natural-only and natural-and-anthropogenic emissions for 2001 have been generated over North America at a 1°×1° resolution using the nested version of the GEOS-Chem model. The resulting monthly-mean AOD values for individual aerosol types within GEOS-Chem grid boxes are then interpolated to provide the corresponding optical thickness values for each RCM grid box and each aerosol type during the course of the model integration. The aerosol types in GEOS-Chem are closely matched with those parameterized in the Fu-Liou-Gu scheme; thus the implementation of the GEOS-Chem aerosol fields in RCM simulations is straightforward.

The RCM simulations have been performed for the period January-December 2001 using the large-scale forcing from the National Centers for Environmental Prediction (NCEP)-Department of Energy (DOE) Reanalysis version 2 (Kanamitsu et al. 2002). The model domain (Fig. 1) covers the WUS region at a 36 km horizontal resolution and 18 atmospheric and four soil layers in the vertical. This domain configuration has been successfully used in a number of previous studies for seasonal water cycle (Kim 1997; Kim et al. 1998, 2000), decadal climate variability (Kim and Lee 2003), and climate change projections (Kim 2001, 2005; Kim et al. 2002) in WUS. These previous studies have shown that MAS run with this domain configuration can capture the characteristics of the regional climate with reasonable accuracy. The two small boxes within the model domain (Fig. 1) are the Northwestern United States (NWUS) and Arizona-New Mexico (AZNM) regions. These two regions have been selected in order to examine the radiative effects of anthropogenic aerosols under contrasting regional climate characteristics within the WUS region; wet winters and dry summers in NWUS and wet winters/summers and dry springs/falls in AZNM (Higgins et al. 1997; Kim 2002; Kim and Lee 2003).

To calculate the effect of anthropogenic aerosols, we have performed four simulations in which the monthly-mean AOD fields in the corresponding CTM runs are prescribed (Table 1). The control run (CTRL) includes the effect of all aerosols of both natural and anthropogenic origins. The natural aerosol run (experiment NAER) utilizes the GEOS-

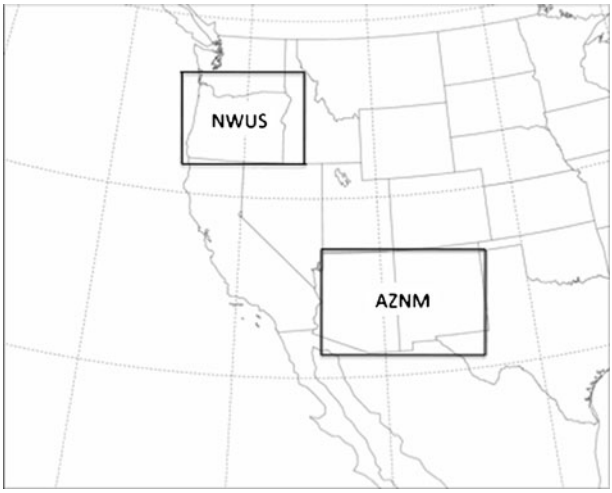

Fig. 1 The WUS domain; the two inner boxes indicate the NWUS and AZNM regions selected for further analysis

Chem aerosol field generated by considering only the emissions inputs corresponding to natural sources. The radiative effect of the total anthropogenic aerosols is then obtained by subtracting the results in NAER from those in CTRL. Two additional RCM runs have been performed to examine the radiative effects of anthropogenic sulfates and BC. For calculating the effect of anthropogenic sulfates, the AOD field used in CTRL is modified in such a way that the AOD associated with the total sulfates are replaced by that with the sulfates of natural-only origins (experiment SULF). Subsequently, the radiative effect of anthropogenic sulfates is calculated as the difference between experiments CTRL and SULF. The effect of anthropogenic BC is calculated in a similar way; the BC AOD field in CTRL is replaced by the AOD field of the natural BC (experiment BLCB). The radiative effect of anthropogenic BC is then examined from the difference between CTRL and BLCB. Note that the aerosol radiative effect calculated in this way from the RCM includes the direct effect and the feedback among interactive components of the model climate system, especially clouds, such as semi-direct effect (e.g., Johnson 2005; Johnson et al. 2006); however, in the absence of the impact on the properties of cloud particles, the aerosol indirect effects are not included. Typical observational estimate of direct aerosol radiative effect is obtained only for cloud-free areas, thus the impact of the feedback through other climate components is not accounted for. Because the direct and semi-direct effects are inseparable within the current RCM structure and the experimental design, the combined direct- and semi-direct radiative effects will be called radiative effects below.

3 Seasonal AOD

The column-integrated AOD of the CTM-simulated anthropogenic aerosols varies according to geography and season (Fig. 2). The evaluation of the GEOS-Chem results used in this study has been presented in Park et al. (2004, 2006) and will not be discussed here. Seasonally, the smallest (largest) AOD of the total anthropogenic aerosols occurs during winter (summer) in most of WUS. This seasonal variability is associated with larger formation of secondary aerosols (Park et al. 2004) and more frequent wildfires (Park et al. 2003, 2007) during summer in WUS. The geographical variations in the AOD during winter are characterized by strong zonal gradients with minimum (maximum) values in the Pacific coast region (to the east of the Continental Divide). This zonal gradient in the AOD occurs in all seasons because the prevailing winds over WUS are generally westerly that bring pristine ocean air into the region, especially during the cold season. In addition, winter is the wettest season in the region to the west of the Continental Divide. Thus, wet scavenging of aerosols by widespread stratiform precipitation during winter results in low aerosol concentration and small AOD. During summer, the prevailing westerlies are

Table 1 The numerical experiments performed in this study

Experiment	GEOS-Chem aerosol fields used in the RCM simulation
Control (CTRL)	GEOS-Chem aerosols from anthropogenic and natural emissions
Natural (NAER)	GEOS-Chem aerosols from natural emissions only
Sulfate (SULF)	All GEOS-Chem aerosols except anthropogenic sulfates
	(CTRL-SULF is related with the presence of anthropogenic sulfates)
Black Carbon (BLCB)	All GEOS-Chem aerosols except anthropogenic BC
	(CTRL-BC is related with the presence of anthropogenic BC)

weakened, especially over AZNM where low-level southerly winds develop in association with the onset of the North American Monsoon (NAM) (Kim and Lee 2003; Kim et al. 2005). This reveals that the southerly winds that are the major source of moisture in AZNM during the monsoon season also transport a large amount of anthropogenic aerosols and aerosol precursors from Mexico (Pitchford et al. 2004). Wet scavenging is also minimal in summer over WUS except in the region affected by the NAM, especially AZNM. The seasonal variations in wind and rainfall result in the summer AOD field that is characterized by more complex spatial patterns than in winter with additional north–south AOD gradients Southwestern US (SWUS).

The seasonal and geographical variations in AOD due to anthropogenic aerosols appear clearly in the seasonal AOD averaged over the two regions, NWUS and AZNM (Fig. 3). The summertime AOD peak in AZNM is much larger than that in NWUS. Summer is a wet season in AZNM (Higgins et al. 1997; Kim 2002; Kim et al. 2005), thus the effect of the cross-border pollutant transport by the monsoon circulation dominates the effect of wet scavenging by monsoon rainfall in determining the summertime AOD field over AZNM, as shown in the GEOS-Chem simulations. The AOD of the entire anthropogenic aerosols is mainly associated with anthropogenic sulfate; BC contributes about 10–15% of the total.

4 Radiative effects of anthropogenic aerosols on radiative transfer

The radiative effect of anthropogenic aerosols on radiative transfer is examined through the comparison of the simulation results between CTRL and NAER. Note again that the term

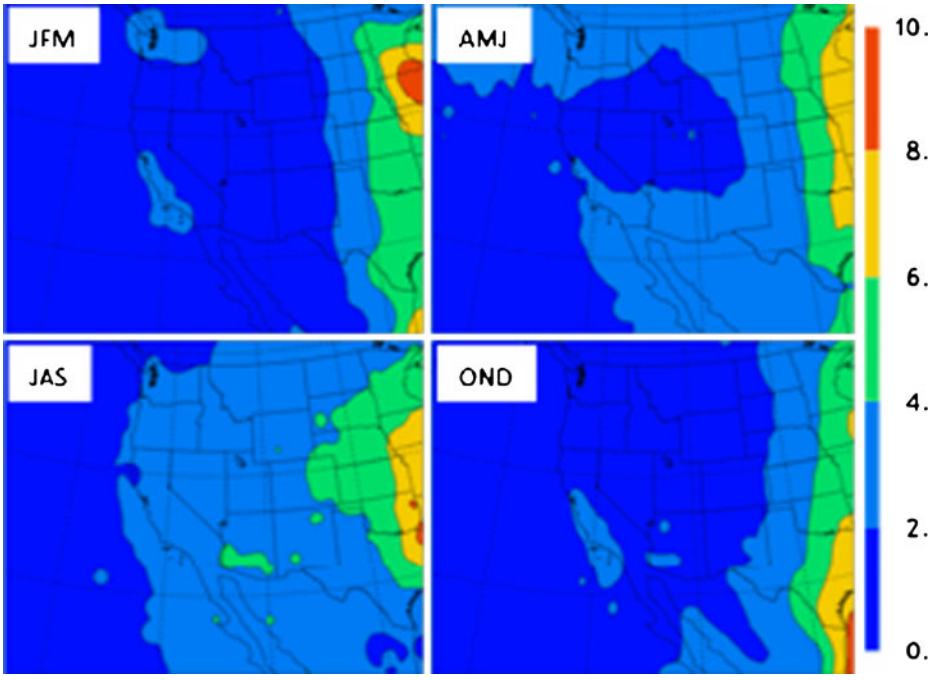

Fig. 2 The simulated seasonal-mean AOD for all anthropogenic aerosols in 2001. The AOD values in the plot have been multiplied by 100

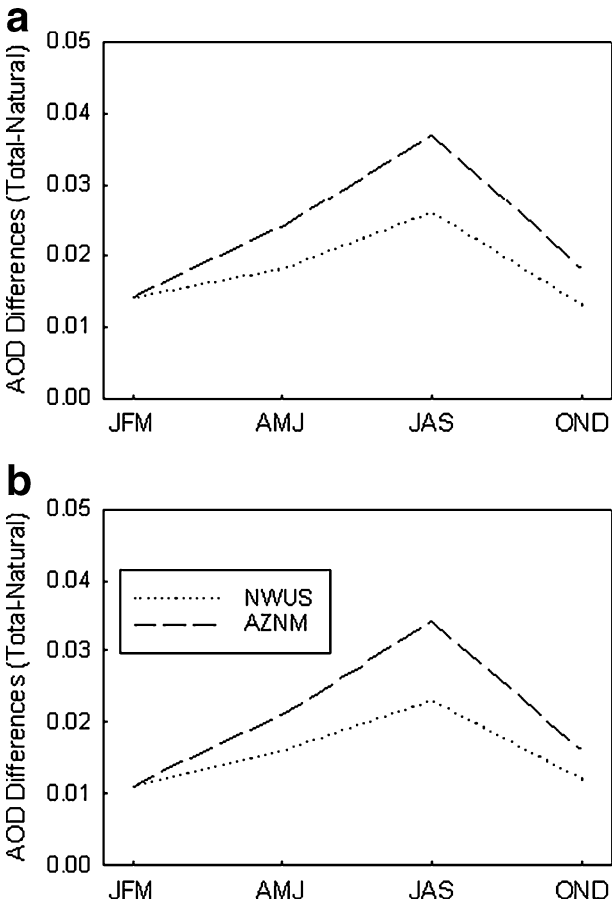

Fig. 3 The seasonal-mean AOD by (**a**) the total anthropogenic aerosols and (**b**) the anthropogenic sulfate in NWUS and AZNM simulated in the GEOS-Chem model

'radiative effect' in this study stands for the combined direct and semi-direct effects for brevity. At the top of the atmosphere (TOA), the changes in the outgoing shortwave radiation (OSRT) due to the anthropogenic aerosols vary according to season and geography. The magnitude of the radiative effect on OSRT by the total anthropogenic aerosols is largest in summer, especially in AZNM and in the northeastern interior part of NWUS, with the local peaks from $-10\,\mathrm{W\,m^{-2}}$ to $+10\,\mathrm{W\,m^{-2}}$ (Fig. 4). Here, OSRT is defined positive upward; i.e., the positive values in Fig. 4 correspond to negative TOA shortwave radiative forcing for the atmospheric column. The radiative effect of the total anthropogenic aerosols on OSRT is positive (i.e., negative shortwave radiative forcing) in most WUS because shortwave-reflecting sulfates dominate. Locally large negative and positive shortwave radiative forcing coexists in AZNM during summer (Fig. 4c) where significant summertime convection occurs. The local negative values, i.e., reduced OSRT, closely coincide with reduced cloud water path (CWP) as shown in the following section. This reveals that the feedback via clouds, i.e., the semi-direct effect, plays an important role in modulating the overall radiative effects on shortwave radiative transfer in the regions and seasons of significant convection. The radiative effect of the total anthropogenic aerosols on OSRT found in this study is smaller than that suggested in previous modeling studies for the heavily polluted regions in China (Giorgi et al. 2002; Huang et al. 2007; Gu et al. 2006)

where the TOA shortwave radiative forcing of −1 to −15 Wm^{-2} by anthropogenic sulfates was obtained. Because the AOD of the total anthropogenic aerosols mostly comes from anthropogenic sulfates, the radiative effect of the total anthropogenic aerosols on radiative transfer and clouds are close to those of anthropogenic sulfates. Thus, only the radiative effects of the anthropogenic sulfates and BC in the NWUS and AZNM regions are further discussed below.

To investigate the seasonal and geographical variations in the effects of anthropogenic aerosols, we examine the seasonal-mean radiative effects by anthropogenic sulfates and BC over NWUS and AZNM, the regions of contrasting climate characteristics. The results show that the radiative effect of anthropogenic sulfates on OSRT varies from 0.2 Wm^{-2} to 1 Wm^{-2} according to season and geography (Fig. 5a). The radiative effect of anthropogenic sulfates on OSRT is positive in both regions. In NWUS, the maximum and minimum effects of anthropogenic sulfates on OSRT occur during summer and winter, respectively. The effect in AZNM shows a bimodal seasonal distribution with spring/fall maxima and winter/summer minima. It is shown that the maximum effect of scattering sulfates normally occurs in dry season when the cloud feedback is much smaller. Anthropogenic BC reduces OSRT, i.e., positive shortwave forcing, in both regions due to substantial absorption of solar radiation, with the maximum effect in summer (Fig. 5b). Thus, the positive effect of anthropogenic BC partially compensates the negative effect of anthropogenic sulfates on OSRT. Consequently, the radiative effect of the total anthropogenic aerosols appears somewhat smaller than anthropogenic sulfates (not shown). Note that the sum of the effects of anthropogenic sulfate and black carbon does not exactly correspond to the effect of the total anthropogenic aerosols. This shows that the overall aerosol radiative effect in the climate system is modulated by nonlinear feedback from the clouds, and that a sum of the

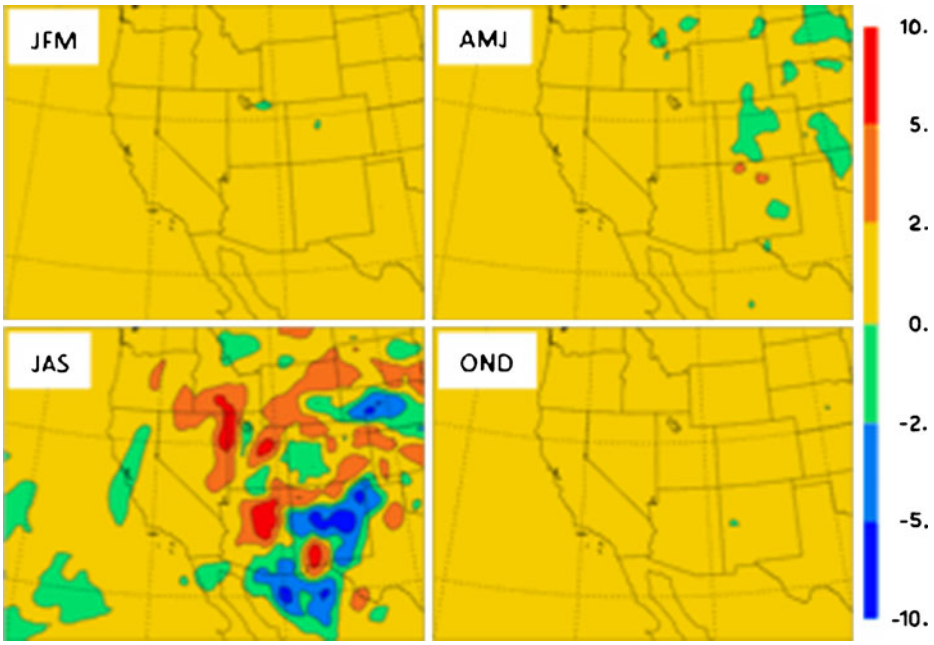

Fig. 4 The seasonal-mean changes in OSRT due to the direct radiative effects of the total anthropogenic aerosols (Wm^{-2})

radiative effects of individual aerosol types cannot be simply used to estimate the overall effect produced by multiple aerosol types.

The radiative effect of anthropogenic sulfates and BC on surface insolation is negative throughout the year (Fig. 6). The effect of anthropogenic sulfates (Fig. 6a) undergoes a seasonal cycle similar to that on OSRT (Fig. 5a). The maximum and minimum effects on surface insolation by anthropogenic sulfates in NWUS are −1.7 and −0.4 Wm^{-2} in summer and winter, respectively. The radiative effect on surface insolation by anthropogenic sulfates in AZNM shows winter/summer minima and spring/fall maxima because the aerosol radiative effects on surface insolation are strongly masked by clouds in the wet season (Kim et al. 2006). The radiative effect on surface insolation by anthropogenic BC (Fig. 6b) in AZNM shows a bimodal seasonal distribution with a maximum (minimum) in the spring/fall (winter/summer), also due to the cloud effect.

Notable amounts of the radiative effect of anthropogenic aerosols on outgoing longwave radiation (OLR) occur only in summer and vary significantly according to geography (Fig. 7). Anthropogenic sulfates increase summertime OLR by 0.8 Wm^{-2} over AZNM and

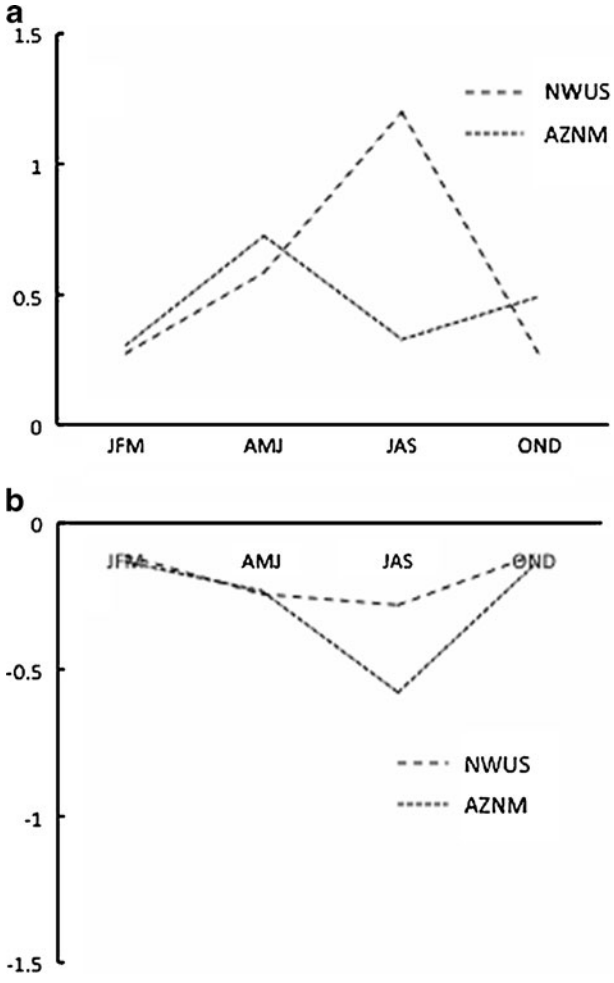

Fig. 5 The radiative effects (Wm^{-2}) on OSRT by (**a**) anthropogenic sulfate and (**b**) anthropogenic black carbon

reduce it over NWUS by 0.4 Wm^{-2} (Fig. 7a). These effects on OLR are related with the decrease of about 0.4 Wm^{-2} in the downward longwave radiation at the surface (DLRS) over AZNM and an increase of 0.3 Wm^{-2} over NWUS (Fig. 8a). Sulfates mainly scatter solar radiation and exert no significant impact on longwave radiation directly (Gu et al. 2006); thus the changes in the OLR and DLRS shown here are mainly through the modification of clouds by anthropogenic sulfates. The magnitude of the radiative effect of anthropogenic sulfates on OLR and DLRS is smaller than that on OSRT (Fig. 5a) or insolation (Fig. 6a) in both regions except in AZNM during summer due to the modification of clouds as will be shown in the following section. The radiative effects of the total anthropogenic aerosols are similar to that of anthropogenic sulfates with +1.14 Wm^{-2} and −0.52 Wm^{-2} in AZNM and NWUS, respectively, in summer (not shown). Anthropogenic BC affects OLR in a similar way as anthropogenic sulfates (Fig. 7b). The radiative effects of anthropogenic BC on OLR are also strongly modulated via the modification of clouds as

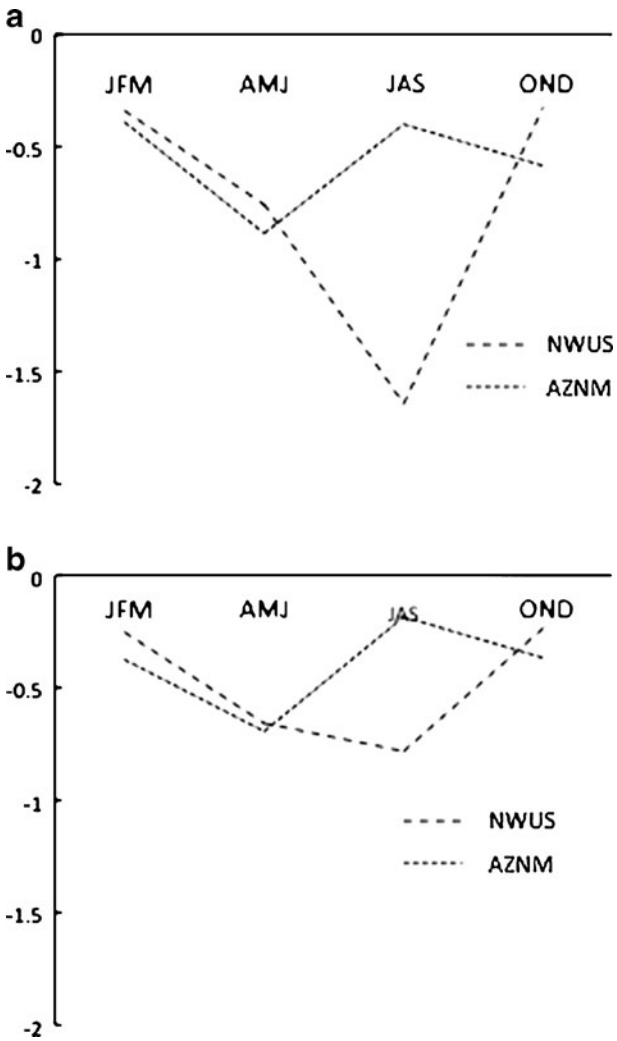

Fig. 6 The radiative effects (Wm^{-2}) on surface insolation by (**a**) anthropogenic sulfate and (**b**) anthropogenic black carbon

discussed in the following section. Anthropogenic BC reduces DLRS in all regions, but noticeable effects occur only in AZNM where DLRS is reduced by 0.33 Wm^{-2} during summer (Fig. 8b).

5 Effect on clouds

The radiative effects of anthropogenic aerosols on clouds are examined in terms of the seasonal mean CWP. Noticeable effects of anthropogenic aerosols on CWP in both NWUS and AZNM occur only during summer when convective clouds are dominant, with much larger impact in AZNM (Fig. 9). Anthropogenic sulfates reduce (−5.1%) the CWP in AZNM but increase (+2.6%) it in NWUS for summer (Fig. 9a). The magnitude of the CWP changes due to the anthropogenic aerosols is smaller than the natural variability measured in terms of the temporal standard deviations. The opposite effects of the anthropogenic sulfates on CWP in NWUS and AZNM may be related with the differences in the summer

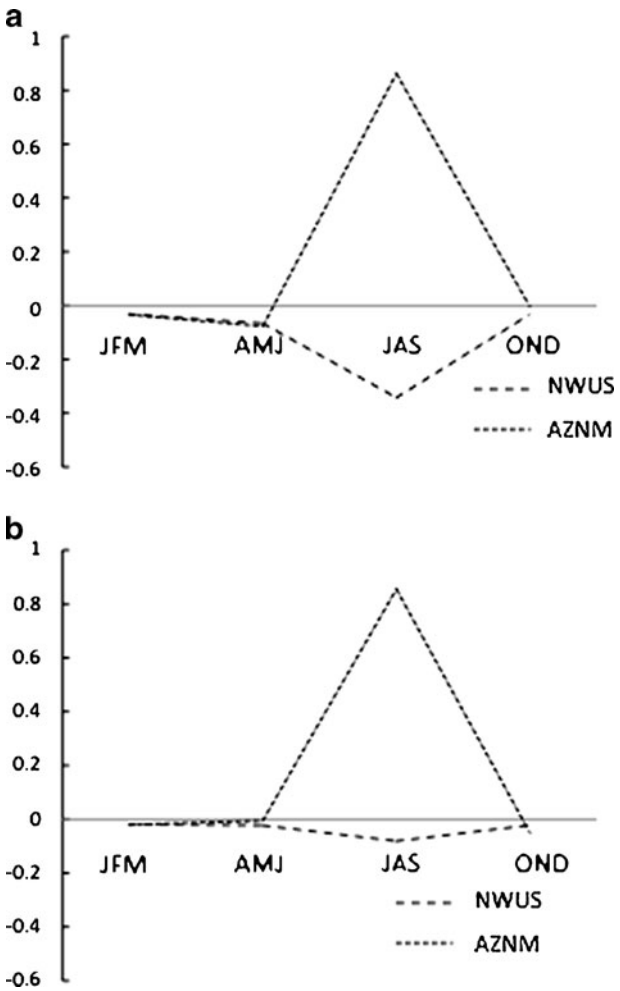

Fig. 7 The radiative effects (Wm^{-2}) on OLR by (**a**) anthropogenic sulfate and (**b**) anthropogenic black carbon

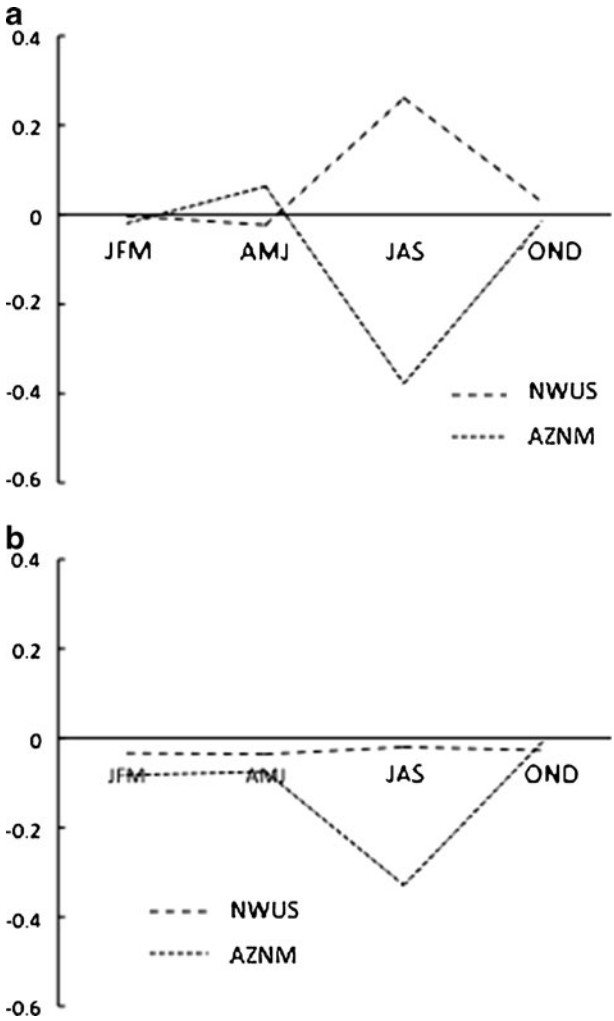

Fig. 8 The radiative effects (Wm^{-2}) on the downward longwave radiation at the surface (DLRS) by (**a**) anthropogenic sulfate and (**b**) anthropogenic black carbon

climate between the two regions. Significant convection occurs in AZNM due to the NAM circulation while non-precipitating clear-sky cumuli are the most frequent type in NWUS during summer. Considering the difference in the type of convective clouds between the two regions, the results show that the effect of shortwave-scattering sulfates tends to reduce cumulus clouds driven by strong convection as found in previous studies (Giorgi et al. 2002; Huang et al. 2007) while they increase non-precipitating clear-sky cumuli driven by local atmospheric instability. The reduced CWP in AZNM partially offsets the shortwave-scattering effect of the anthropogenic sulfates on the summertime surface insolation and OSRT. The changes in CWP can also be related with the changes in OLR and DLRS because larger CWP traps more OLR. Thus, the increase and decrease in OLR in AZNM and NWUS (Fig. 7a), respectively, can be related with the changes in CWP due to the anthropogenic sulfates (Fig. 9a). The changes in CWP can be similarly related with the decrease and increase in the summer DLRS over AZNM and NWUS, respectively, due to the anthropogenic sulfates (Fig. 8a).

The effects of anthropogenic BC on CWP is negative in both regions (Fig. 9b), and is more significant in AZNM during summer because the warming due to the absorption of solar radiation reduces clouds generated by the monsoon circulation (Menon et al. 2002; Gu et al. 2006; Huang et al. 2007). The reduced CWP results in decreased OSRT (Fig. 5b), increased OLR (Fig. 7b), and decreased DLRS (Fig. 8b). The reduced surface insolation (Fig. 6b), however, is associated with the absorption of solar radiation by anthropogenic BC that exceeds the cloud feedback effect produced by the reduced CWP. Overall, the seasonal and geographical variations in the radiative effects of anthropogenic aerosols on the radiation budget are related with the regional climate characteristics, in particular, the amount and type of the dominant clouds, because clouds can strongly mask the direct aerosol radiative effects (Kim et al. 2006). Details of the physical processes, for example the aerosol-induced diabatic heating and the corresponding alterations in static stability, behind the aerosol-cloud feedback found in this study could not be analyzed due to our inability of tracking the complex interaction and feedback within the model. This will be left for future studies.

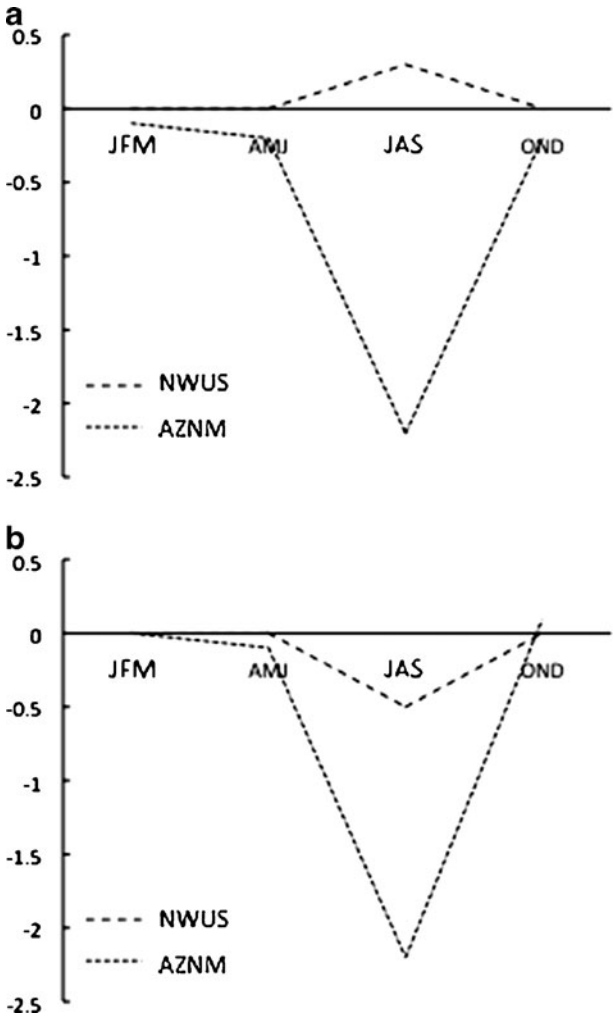

Fig. 9 The radiative effects (fraction of the amount in the control run) on the column-integrated CWP by (**a**) anthropogenic sulfate, and (**b**) anthropogenic BC

6 Conclusions

We have examined the combined direct and semi-direct radiative effects of anthropogenic aerosols on the radiative transfer and clouds in WUS using an RCM in conjunction with the aerosol fields simulated in the GEOS-Chem model. Two sets of monthly-mean AOD fields corresponding to natural-and-anthropogenic and natural-only emissions have been generated using GEOS-Chem. The CTM aerosol datasets have been implemented in four RCM simulations to compute the radiative effect of anthropogenic aerosols on the radiative transfer and CWP in two regions, NWUS and AZNM, of contrasting climate characteristics.

The CTM results show that the geographical and seasonal variations in AOD by anthropogenic aerosols are determined primarily by local emissions and regional climate, especially the prevailing winds and precipitation. The AOD associated with anthropogenic aerosols is largest in summer and smallest in winter in both regions. During the winter, prevailing westerly winds and significant precipitation result in strong zonal gradients in AOD with the minimum and maximum values occurring in the Pacific coastal region and to the east of the Continental Divide, respectively. In summer, the low-level southerly winds associated with the development of North American monsoon results in large AOD values over SWUS due to the trans-border pollutant from Mexico. The entire anthropogenic aerosols behave primarily as shortwave-reflecting aerosols because their AOD is mostly associated with anthropogenic sulfates with only a minor contribution from the anthropogenic black carbon.

The direct radiative effect of the entire anthropogenic aerosols on OSRT is characterized by significant geographical and seasonal variations, especially during summer in SWUS and the inland region of WUS where their local magnitude varies from -10 Wm^{-2} to $+10$ Wm^{-2}. The radiative effect of anthropogenic sulfates increase OSRT by 0.2 to 1 Wm^{-2}, thus negative shortwave radiative forcing at TOA, in both NWUS and AZNM. In NWUS, the maximum and minimum effects on OSRT occur in the summer and the winter, respectively. In AZNM, the radiative effect on OSRT is characterized by a bimodal seasonal distribution with the maxima (minima) in spring and fall (winter and summer). The radiative effect of anthropogenic black carbon reduces OSRT, thus positive TOA shortwave radiative forcing, for all seasons with the maximum effect during summer in both regions. The radiative effect of the entire anthropogenic aerosols on OSRT closely resembles that by anthropogenic sulfates. The effect of anthropogenic sulfates on surface insolation that is negative in all regions undergoes similar seasonal and geographical variations as their effect on OSRT. The seasonal variations in the effect of anthropogenic BC on surface insolation in AZNM show a bimodal seasonal distribution with the maxima in spring and fall and the minima in winter and summer.

The radiative effects of anthropogenic aerosols on OLR and DLRS are noticeable only during summer. Both anthropogenic sulfates and black carbon increase (decrease) OLR in AZNM (NWUS). The radiative effect of anthropogenic aerosols on the summer DLRS in NWUS is opposite to that in AZNM; both anthropogenic sulfates and black carbon decrease (increase) DLRS in AZNM (NWUS).

The radiative effect of anthropogenic aerosols on CWP is noticeable only during summer when convective clouds are significant as well. Anthropogenic aerosols reduce CWP in AZNM, but slightly increase it in NWUS during summer. This shows that the radiative effect of anthropogenic aerosols on CWP depends on regional climate characteristics in which convective clouds develop. They reduce significant

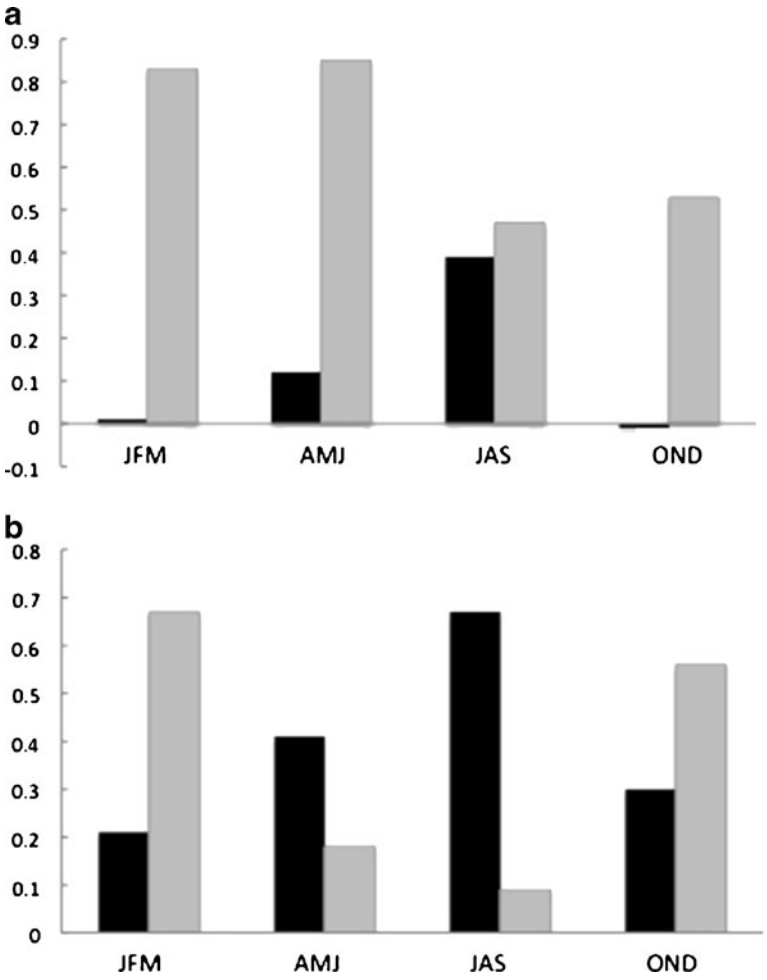

Fig. 10 The spatial correlation coefficient between the direct radiative effects of anthropogenic sulfates on OSRT and CWP (*black*). *Gray bars* are the correlation coefficients between the radiative effects and the AOD of anthropogenic sulfates: (**a**) NWUS, (**b**) AZNM

convection driven the NAM circulation (summertime in AZNM), but enhance non-precipitating clearly-sky cumuli driven by local atmospheric instability (summertime in NWUS).

The examination of the correlation coefficients between the radiative effects on OSRT and two fields, AOD and CWP, reveals that the feedback through clouds is crucial in determining the aerosol radiative effect (Fig. 10). The effect of the anthropogenic aerosols on shortwave radiation is more closely correlated with AOD during winter, but with the changes of CWP during summer in both NWUS and AZNM regions. The results show that the effect of the cloud feedback is particularly important in climate conditions where convective clouds are dominant. Details of the associated physical and dynamical processes are a subject of future research.

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