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Aerosol Microphysical and Radiative Effects on Continental Cloud Ensembles

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Abstract

Aerosol-cloud-radiation interactions represent one of the largest uncertainties in the current climate assessment. Much of the complexity arises from the non-monotonic responses of clouds, precipitation and radiative fluxes to aerosol perturbations under various meteorological conditions. In this study, an aerosol-aware Weather Research and Forecasting (WRF) model is used to investigate the microphysical and radiative effects of aerosols in three weather systems during the March 2000 Cloud Intensive Observational Period campaign at the Southern Great Plains site of the US Atmospheric Radiation Measurement Program. Three cloud ensembles with different meteorological conditions are simulated, including a low-pressure deep convective cloud system, a series of less-precipitating stratus and shallow cumulus, and a cold frontal passage. The WRF simulations are evaluated by the available observations of cloud fraction, liquid water path, precipitation, and surface temperature. The microphysical properties of cloud hydrometeors, such as their mass and number concentrations, generally show monotonic trends as a function of cloud condensation nuclei concentrations. Aerosol radiative effects

do not interfere the trends of cloud microphysics, except for the stratus and shallow cumulus cases where aerosol semi-direct effects are identified. The precipitation changes by aerosols vary with the cloud types and their evolving stages, with more prominent aerosol invigoration effect and associated enhanced precipitation from the convective sources. The simulated aerosol direct effect suppresses precipitation in all three cases but does not overturn the direction of precipitation changes by the aerosol indirect effect. Cloud fraction exhibits much smaller sensitivity (typically less than 2%) to aerosol perturbations than the cloud microphysics, and the responses vary with aerosol concentrations and cloud regimes. The surface shortwave radiation shows a monotonic decrease by increasing aerosols, while the magnitude of the decrease depends on the cloud type. Surface temperature changes closely follow the modulation of the surface radiation fluxes.

1. Introduction

The influence of atmospheric aerosols on the energy budget and hydrological cycle remains one of the least understood aspects in the earth system. Aerosols, from both natural and anthropogenic sources (Zhang et al., 2004; Levy et al., 2013), directly scatter and absorb incoming solar radiation, which alter vertical atmospheric temperature structure, surface and top-of-atmosphere (TOA) radiation fluxes, and cloud fraction (Ackerman et al., 2000; Fan et al., 2008). The large uncertainty in the aerosol direct effect is related to the particle size, chemical composition, and mixing state (Khalizov et al., 2009; Zhang et al., 2015; Peng et al., 2016). Depending on those complicated factors, aerosols impose a net positive or negative forcing over different regions. Presently, the aerosol direct forcing is estimated with a global mean of -0.27 W m^{-2} (IPCC, 2013). In addition to the direct radiative effect, absorbing aerosols have a positive feedback that reduces cloud coverage, namely the semi-direct effect (Hansen et al., 1997; Johnson et al., 2004). The presence of absorbing aerosols in a given vertical layer decreases the atmospheric instability and reduces ambient relative humidity through diabatic heating of the air. This, in turn, enhances cloud evaporation and inhibits convection and vertical mixing, thereby hindering cloud formation, reducing radiative cooling at the TOA and eventually leading to a positive radiative forcing (Ackerman et al., 2000; Wang et al., 2013a).

By acting as cloud condensation nuclei (CCN) or ice nuclei (IN), aerosols affect the micro- and macro-physical properties of different types of clouds, impacting their radiation, dynamics, precipitation, and lifetime. The first indirect effect is primarily related to the impact of aerosols on the cloud droplet size and number (Twomey, 1977;

Albrecht, 1989; Zhang et al., 2007; Yuan et al., 2008). It has been widely accepted that through the first indirect effect, higher concentrations of aerosols lead to higher concentrations of CCN and cloud droplets. Changing the number and size distribution of cloud droplets due to aerosols consequently alters the vertical depth and lifetime of clouds as well as precipitation processes, which is commonly known as the second indirect effect (Albrecht, 1989; Pincus and Baker, 1994; Fan et al., 2007a, b). By changing the size distribution, the growth of cloud droplets by collision/coalescence becomes suppressed, which reduces drizzle and prevents the loss of cloud water content, leading to an increased cloud lifetime (Albrecht, 1989; Rosenfeld, 1999). For certain type of cloud like trade wind cumulus, the reduced precipitation results in deeper cloud layer but smaller cover due to the stronger evaporation (Seifert et al., 2015). Hence such an effect buffers the aerosol lifetime effect (Stevens and Feingold, 2009). In mixed phase clouds, polluted conditions suppress warm-cloud processes but enhance convective development through the aerosol invigoration effect (Koren et al., 2010; Tao et al., 2012; Wang et al., 2011; Fan et al., 2012), leading to enhanced cloud electrification and lightning activities (Williams et al., 1991; Nesbitt et al., 2000; Orville et al., 2001). A complication is that since the cloud particle sizes also increase in a stronger convection (Jiang et al. 2011), the aerosol induced convective invigoration likely balances the reduction of cloud particle sizes due to the Twomey effect. The second indirect effect varies with the cloud type and ambient conditions, such as relative humidity, vertical wind shear, and convective potential energy (Tao et al., 2012; Fan et al., 2016). In cold clouds, aerosols act as IN and increase the number of ice crystals (Sassen et al., 1995; Strom and Ohlsson, 1998). By the Wegener-Bergeron-Findeisen process, ice crystals

grow at the expense of liquid droplets through vapor deposition (Rogers and Yau, 1989), as well as through the processes of riming, aggregation, and accretion. It is still uncertain whether this leads to an enhanced greenhouse effect or a reduction of solar radiation by brighter clouds (Sassen et al., 1995; Lee et al., 2009a).

The impact of aerosols on precipitation and cloud macrophysics is even more complex and diverse, representing the least understood component of the aerosol effects (Koren et al., 2013; Rosenfeld et al., 2014). Recent studies suggest that the impact of aerosols on precipitation depends on the type of clouds and the environmental conditions, in which clouds form (Khain, 2009; Li et al., 2011; Lin et al., 2016). In addition, Li et al. (2008) showed that the aerosol effect is non-monotonic for a convective cloud, i.e., an initial enhancement at low aerosol concentrations, but a suppression at high aerosol concentrations for precipitation, updraft speed, and cloud fraction. Tao et al. (2007) also showed a switch of aerosol effects on convective clouds, from suppression to enhancement, in different locations.

It is critical to examine the aerosol effects on different cloud types under various weather systems. Numerous previous modeling studies have been devoted to understanding the physical mechanisms of the aerosol effects. However, most of those studies only deal with either the aerosol direct or indirect effect and focus on a certain cloud type. This study aims to investigate both aerosol radiative and microphysical effects jointly and individually and to quantify the overall aerosol effects on different cloud regimes. Another objective of this modeling study is to explicitly assess the monotonicity in the aerosol-cloud-radiation relationships. Specifically, this study explores the monotonicity of cloud responses to aerosol variations under different cloud

regimes, which has profound implications for parameterizations of aerosol-cloud relationships in global climate models.

2. Experiment Setup

2.1 Model Description

In this modeling study, the cloud-resolving Weather Research and Forecast (WRF) model version 3.1.1 is used. A two-moment bulk cloud microphysical scheme and a modal aerosol scheme were implemented (Li et al. 2008; Wang et al., 2011) to account for the aerosol-cloud-radiation interactions. The two-moment bulk cloud microphysical scheme includes the mass mixing ratio and number concentration for five hydrometeor types—cloud droplets, raindrops, ice crystals, snow, and graupel. The size distribution for each of the five hydrometeors is determined via the gamma function (Li et al., 2008), and thirty-two microphysical processes are considered including an explicit condensation calculation using supersaturation and an autoconversion scheme based on relative dispersion (Liu and Daum, 2004). There is no chemistry component in our model.

The Goddard radiation scheme was modified (Fan et al., 2008; Wang et al., 2014b) to include the radiative forcing by the aerosol direct/semi-direct effects. The aerosol module determines aerosol radiative properties, including optical depth, asymmetry factor and single scattering albedo, as a function of wavelength, composition, mixing state, and relative humidity. To enhance the computational efficiency, a lookup table is developed for the optical properties for all size ranges once the aerosol radiative properties are determined.

For each model run, three nested two-way domains are used (Figure 1a) with 18, 6, and 2 km spatial resolution and a 12 second temporal resolution. The innermost

domain is roughly 350 km by 350 km with 50 vertical levels and centered at (36.6°N, 97.5°W), covering the same domain as the US Department of Energy Atmospheric Radiation Measurement (ARM) Southern Great Plain (SGP) site near Ponca City, OK. All model data analyzed in this study are from the innermost 2-km domain with 15-minute output intervals. The North American Regional Reanalysis data is used for the initial and boundary meteorological conditions.

2.2 Aerosol Profiles

To study the aerosol direct and indirect effects and to obtain the monotonicity of the cloud responses, three different aerosol profiles are utilized in this study to represent clean, moderate and polluted environments, respectively (Figure 1b). Two types of anthropogenic aerosols mainly considered in this study are sulfates and black carbon. The clean cases use a relatively clean background continental profile with an initial surface aerosol number concentration of 210 cm^{-3} , as used by Li et al. (2009) from the Texas Air Quality Study (TexAQS) 2000 campaign. The moderate cases, herein referred to as SGP cases, are based on aerosol measurements taken at the SGP site during the 2003 Aerosol Intensive Observation Period (IOP) campaign. The SGP cases have an initial surface concentration of 1200 cm^{-3} . The polluted cases are 10 times the SGP profile with an initial surface concentration of 12000 cm^{-3} , which is a similar magnitude as the urban case used by Cheng et al. (2007). For all three profiles, the aerosol concentration is assumed to decrease exponentially with height above about 5 km (Cheng et al., 2007; Li et al., 2009). As aerosol mass and number concentrations are prognostic variables in the model, aerosol vertical mixing and horizontal advection are explicitly considered. There is no direct emission of aerosols from the surface, but aerosols can be advected into the

inner domains from the boundaries under favorable wind conditions. Aerosols are removed by nucleation scavenging. Internal mixing of 95% sulfate and 5% black carbon by mass is assumed for studying radiative effects of light-absorbing aerosols. Such a composition assumption for radiation calculation has been justified by previous field measurements (Levy et al., 2013) and modeling study (Wang et al., 2014b) in Southern US. Each aerosol profile will be used for the simulations with the aerosol direct and indirect effects (DIE) and the simulations with the aerosol indirect effect only (IEO).

2.3 Case Studies

From March 1 to March 26, 2000, the Cloud Intensive Observation Period (IOP) campaign was conducted at the ARM SGP site. The goal of the campaign was to collect three-dimensional cloud properties from observational data including the standard set of ARM SGP instruments, radar and lidar observations, and aircraft measurements, which included a total of 12 flights during the period. The cloud data has been divided into 6 subperiods (A through F) that contain different synoptic and cloud properties and have been extensively studied regarding cloud-climate feedback in atmospheric general circulation models (Zhang et al., 2005). For this study, aerosol-cloud interactions will be studied for three of the six subperiods (hereafter denoted as A, D, and E for convenience).

Case A pertains to clouds to the north of a developing low-pressure system from 1500Z on March 1 to 0000Z on March 5 (Figure 2a). About 10 hours prior to the start of this case, a cold front moved through the domain at about 0430Z on March 1. Clouds began to move into the domain from a low pressure system that formed in the Four Corners region around 2000Z on March 1. By 2000Z on March 2, the low pressure system entered the southwest corner of the domain while a thick layer of clouds covered

the region. Between March 2-3, the center of the low moved along the Oklahoma-Texas boarder with cloud development primarily to its north. By 1330Z on March 3, the system left the region with the skies mainly clearing up by 1200Z on March 4 as a high-pressure system kicked in.

Case D pertains to a collection of less-precipitating clouds, i.e. a series of stratus from 2100Z on March 11, 2000 to 1200Z on March 14 (thereafter referred as Case D1) and a shallow cumulus from 1200Z on March 14, 2000 to 1200Z on March 15 (Case D2). Prior to the start of this case, there was a stationary front to the south draped across central Texas from the Louisiana-Missouri boarder across to New Mexico, with a high pressure center behind it located centrally over the domain. During March 13, a cold front passed to the north of the domain which also moved the high pressure center out of the region. During March 14, a weak low-pressure system passed through southern Texas, facilitating some shallow convection and light precipitation in Oklahoma (Figure 2b). To better understand the aerosol effects on different types of cloud, our future analyses will be conducted on the two periods separately.

Case E pertains to clouds associated with the genesis of a cold front that moved through the domain from 0900Z on March 15 to 0000Z on March 20, 2000 (Figure 2c). The cold front approached from the north-northwest and arrived in the domain at about 0100Z on March 16. Satellite imagery shows convective development directly over the SGP central facility from 0000Z to 0300Z. By 1300Z the cold front had moved south into Texas and out of the domain where it stalled. Behind the cold front, mainly lower level clouds persisted over the domain due to an upper level low. On March 18, another low

pressure system passed to the south of the domain right long the Oklahoma-Texas boarder. The region finally cleared out by about 2000Z on March 19.

3. Results and Discussion

3.1 Microphysics properties of hydrometeors

Distinct microphysical responses to aerosol initial conditions are shown for all the three cases and vary with the microphysical parameters of interest and depend on cloud types. No matter the heavy-precipitation cases (Case A and Case E) or the case dominated by less-precipitating clouds (Case D), our IEO simulations show that the all of cloud droplet microphysical properties, including mass mixing ratio, number concentration, and effective radius, monotonically change with aerosols, indicating the relative straightforward role of CCN in determining the cloud microphysical properties. Regardless of whether the aerosol radiative effect is included or not, the number concentration of cloud droplets strictly increases as a function of CCN available for water vapor condensation in each case (Figure 4a,d,j,i), and such increase rates are about the same with the aerosol elevation rate we impose in the experiment. The cloud droplet mass content also monotonically increases along with the elevation of aerosol concentration, except for the stratus clouds in Case D1 when aerosol radiative effects are included (Figure 3d). The significant reduction of droplet mass concentration by aerosol radiative effect is found in Case D1. Overall, inclusion of aerosol radiative effects in simulations (DIE cases) has limited influence on the trend of cloud droplets but shows some impacts on the magnitude, particularly for the less-precipitating-cloud dominant case (Case D).

To understand the aerosol radiative influence, we show the air temperature changes under different aerosol conditions between DIE and IEO in Case D (Figure 4). With an increase of aerosol concentration from Clean to Polluted, the variations of air temperature due to aerosol radiative effects become larger. In the polluted condition (Figure 5c), it is clear that there is a significant warming in the lower free troposphere from Day 72 to 74. It can further induce the reduction of relative humidity and decrease the cloud content shown in Figure 3, which is the canonical semi-direct effect. Meanwhile, the warming in the free troposphere and the cooling inside the boundary layer form a temperature inversion and inhibit convection and vertical mixing. Such a thermodynamical effect also contributes to cloud reduction when the aerosol radiative effects are considered in DIE. Another interesting phenomenon is that the altitude of the warming center is elevated from noon to afternoon, indicating the aerosols are lifted upward along with the heated air parcel in the non-precipitating environment.

Both mass mixing ratio and number concentration of raindrops exhibit a decreasing trend in response to aerosol for all the three cases (Figures 3 and 4). This is attributed to the smaller cloud droplets in the polluted conditions, which are not favorable for their collision/coalescence to produce raindrops. Figure 6a-c shows the cloud droplet effective radii are reduced at all cloud points within the cloud ensembles in all three cases from the clean to the polluted condition. The size of raindrops is one of the key controls for the precipitation amount at the surface, but its response to aerosol perturbation is more complicated than those from raindrop mass and number concentrations. By only considering CCN effect in IEO, the increase in the raindrop size is found for most times and levels in three cases (Figure 6d-f), but some scattered reduction in raindrop size

occurs possibly due to the changes in ice-phase particle size like snow and graupel as well as the convective strength.

Different from the cloud droplet and raindrop, ice particles exhibit some non-monotonic responses in different cloud regimes, especially for the less-precipitating stratus and cumulus. Relative to the clean profile, the mass mixing ratio of ice crystals is reduced in the polluted profile by 7% and 40% for Case A and Case E, respectively. The number concentration of ice crystals generally increases as aerosol increases (Figure 4), so their effective radius decreases by varying degrees. The vertical profiles of the ice particle size change show most of radius reduction occur in the upper part of the cloud systems (Figure 6g-i), while lower level ice crystals can even grow bigger at the expense of more supercool droplets. Similar with cloud and raindrops, the aerosol radiative effect in DIE does not change the trend of ice crystals in response to aerosol concentration increases, but it does alter the absolute concentrations of the hydrometers for the two-precipitating cases (Case A and E) compared with IEO. In particular, the number concentration of ice crystal which is greatly enhanced in DIE simulations at high aerosol levels in Case A (Figure 4c). For the stratus in D1, the CCN effects on ice mass and number concentrations are either saturated or reversed from the moderate to heavy polluted conditions. By comparison of DIE and IEO in Figures 4 and 5, it appears that both the monotonicity of microphysical response and the magnitude of microphysics of ice crystals could be greatly modulated by aerosol direct effects in Case D.

3.2 Precipitation

Figure 7 presents the evolution of rain rates throughout Case A. The simulated temporal variation of rain rates generally agrees with the observations based on rain

gauge data from the Arkansas Red-Basin River Forecast Center (ABRFC). For example, both simulations and observations show intensive precipitation started from 0900Z Day 62 and ended around noon time of Day 63. However, the primary peak during the first precipitation period is delayed by half day in the simulations, possible due to the bias in the simulated storm center. The strongest updrafts are observed during the first period, suggesting a convective origin of precipitation. For both the DIE and IEO cases, aerosol concentration and accumulated precipitation are positively correlated, consistent with the larger size of raindrops in more polluted conditions. The larger raindrops could have higher possibility to survive in evaporation when they precipitate out from clouds, leading to more surface precipitations. There is a statistically significant difference between rainfall amounts with and without the direct effect. More precipitation is produced in the IEO case than in the DIE case for both the clean and polluted profiles, which indicates that through blocking more radiation into the atmosphere, the aerosol direct effect systematically weakens the convection strength associated with the cloud development (Figure S1).

Mid-level non-precipitating clouds are primarily present during most period of Case D, but some shallow cumulus clouds were formed to the south of the SGP near the end of the period Central, bringing a brief period of light precipitation, based on the ABRFC observations (Figure 7c). Our model produces some light precipitation at different times during the “non-precipitation” stratus D2 stage according to the observations. The intensive precipitation in the simulations also comes from the convective event in D2, but the rain rates are generally overestimated by a factor of two compared to the observed values. Both observations and simulations show that the

convective event is the only period with significant rainfall and is the main contributor to the accumulated rainfall, so the total accumulated rainfall responses in Figure 7d reflects the changes in D2. Both the DIE and IEO cases overall show similar non-monotonic precipitation trends in response to aerosol, i.e., the accumulated precipitation is enhanced from Clean profile to SGP profile and then is further reduced in the polluted profile. The nonlinearity of precipitation to aerosol can be explained by the corresponding nonlinearity of microphysics effects of aerosol on shallow cumulus clouds. At the relatively low aerosol levels (from clean to SGP), the mass mixing ratio of raindrops decreases by 10% (Figure 3h), which is much smaller than their 58% reduction in number concentration (Figure 4h). It leads to a relative increase in effective radius of raindrops, Hence, raindrops have higher chance to survive through the undersaturation below clouds and results in an enhancement in surface precipitation from Clean profile to the SGP profile. However, due to the larger amount of smaller cloud droplets, warm rain is suppressed in the polluted profile by less efficient collision/coalescence processes. Significant differences in magnitudes of precipitation exist between DIE and IEO cases at high aerosol levels (SPG and polluted). The largest difference between DIE and IEO model runs comes in the polluted profile in the IEO case, which has 43% more rain than the DIE case in average. The higher amounts of precipitation in IEO cases can be traced to higher amounts of cloud, rain, and ice water in the IEO than in the DIE (Figure 3), and it is also consistent with the weaker convection during the period with shallow cumulus cloud in the DIE (Figure S2). By including the direct effect in the DIE cases, absorbing aerosols may reduce the instability by slightly warming the atmosphere, thereby reducing cloud and precipitation formation. It is evident in Figure 4 that, by contracting DIE and

IEO cases, the temperature changes are positive in the atmosphere during daytime and are negative at the surface, suggesting that absorbing aerosols in DIE cases considerably warm the atmosphere and cool the surface correspondingly, thereby weakening the convection, reducing the atmospheric instability, and suppressing cloud and precipitation formation.

During Case E, the model does relatively well in predicting the timing of four maxima of precipitation in comparison to observational data but somehow underestimates the precipitation amount before Day 78 and overestimates it after Day 79 (Figure 7e). The first precipitation maximum was from about 0000Z to 1200Z on Day 76 corresponding to the strongest period of convection associated with the passage of the cold front. Precipitation during this period shows a non-monotonic response to initial CCN concentrations, as the peak values of rainfall rates during this period are 1.27, 1.31, and 1.13 mm hr⁻¹ for clean, SGP, and polluted aerosol profiles with IEO, respectively. Such a non-monotonicity is also found in the CCN effects on a cumulus cloud by Li et al (2008). The next two precipitation maxima occurred during the stratiform rain event from about 1200Z on Day 76 to 1000Z on Day 78, throughout which very few ice particles were present (Figure S3) and there is generally a linear relationship between aerosol concentration and rain rates. The final two precipitation maxima, from about 1200Z on Day 78 to 1400Z on Day 79, are associated with another convection event. The response of rainfall rates to aerosol during this period is a little complicated, as the last precipitation maxima on Day 79 are 0.75, 0.64, and 0.72 mm hr⁻¹ from the clean, SGP, to polluted aerosol profiles. The largest contributor to accumulated precipitation for Case E is due to convective sources on Day 76, hence the overall trend of precipitation in

response to aerosols is first-increase-then-decrease. When comparing accumulated precipitation between DIE and IEO cases, statistically more precipitation (about of 1 ~ 2 mm in average) is produced in the IEO cases than in DIE cases. The higher precipitation for the IEO case could be attributed to the larger amounts of rain and ice water available.

3.3 Cloud Fraction

Figure 8 shows the simulated and observed cloud fraction defined as the fractional area percentage of clouds in an atmospheric layer. Observations were obtained at the SGP Central Facility using the Active Remote Sensing of Clouds Value-Added Product. The modeled cloud fraction is averaged over the 25 nearest grid points (a five-by-five horizontal box around the SGP Central Facility) with a total water mixing ratio (Q_{tot}) greater than $10\text{e-}6 \text{ kg kg}^{-1}$ (Cohen and Craig, 2006), where Q_{tot} is the sum of cloud, ice, snow, and graupel water mixing ratios.

In Case A, as a developing low pressure system passed to the south of the domain along the Oklahoma-Texas border, a few different cloud types, primarily cumulus and stratus clouds, passed through the domain. The general evolutions of cloud fraction are comparable between the simulations and the observation with two distinct periods of deep convection and a long period of low-level clouds. The DIE induces an increase in cloud fraction in both convective clouds and stratiform clouds from clean to polluted conditions (Figure 8a). The overall cloud fraction during the entire case in Figure 8 shows that with the elevated aerosol concentrations, cloud fraction has a fractional increase of 16.6% for DIE and a decrease of 4.6% for IEO. The non-monotonic responses of cloud fraction to different aerosol concentrations in the IEO of this case reveal that cloud fraction may not be a good indicator of the aerosol invigoration effect discussed in

the previous sections. The cloud fraction changes due to aerosols can be largely buffered by the interactions between clouds and ambient air, as the entrainment rate can be modulated after the stronger convection along with the aerosol invigoration effect. One good example is the reduction of relative humidity for the trade wind cumulus in the subtropics (Seifert et al., 2015). Those competing factors make cloud fraction change highly mutable after the aerosol perturbation.

Most of the clouds in Case D are mid-level non-precipitating clouds, some of which contain ice particles. Near the end of the case, some shallow cumulus clouds occurred to south of the SGP Central Facility. Observations show there were primarily two periods of non-precipitating clouds around 0000Z on Day 73 and 74 and a short lived convective cloud late in Day 74 (Figure 8). The simulated clouds in two periods by the model were generally larger and thicker than the observed. The cloud fractions in six experiment runs show large resemblance (Figures 8). The major differences are the reduction of cloudiness at about 0700Z on Day 73 and at about 1800Z on Day 74. Only the aerosol radiative effect emerges in both D1 and D2 showing a negative correlation between aerosol and cloud amount, while the changes in cloud fraction in IEO are insignificant in D1. Cloud fraction can be reduced by 20% due to aerosols in DIE of Case D2.

Cloud cover was continuously present over the SGP domain during Case E primarily due to a cold front, two-day trailing stratiform clouds, and another low-pressure system in the last day. Generally, the model produces thinner cloud depth but consistent evolution compared to the observations (Figures 8). The lower simulated cloud fraction was partly due to the exclusion of rain water mixing ratio in calculating cloud fraction.

Actually, when rain water mixing ratio was included, the modeled cloud fraction appeared exceptionally thicker than observed. It is the same reason for the biased liquid water path (LWP) simulations in the next section. The overall cloud fraction throughout Case E increases with higher aerosol concentrations for both the DIE and IEO cases (Figure 8), with a relative increase of 14.8% and 23.8%, respectively. For the DIE cases, during the cold frontal passage on Day 76, with increased aerosol concentrations, there is an increase in lower level clouds while a decrease in deep convective clouds is observed. During the Day 79 low-pressure system period, the cloud fraction significantly increases. For the IEO cases, with respect to increasing aerosol concentration, cloud fraction increases during each of three periods of note. Also during all three periods, IEO has a larger cloud fraction increase than that in DIE.

3.4 Liquid Water Path

The response of LWP to CCN perturbations is directly related to the aerosol indirect forcing, but the simulated relationships from different climate models do not converge (Fan et al., 2016). The semi-direct effect of absorbing aerosols further complicates the relationship between aerosols and LWP (Lin et al., 2016). In both Case A and E, the simulated LWP exhibits a robust monotonic increase along with the elevation of aerosol loading, due to the CCN effect. Comparing the clean and polluted scenarios, LWP can be enhanced by 50% to 75% in Case A and E. Such a relationship is not subject to the aerosol radiative effect for convective cloud regime, as LWP changes are about the same between DIE and IEO for Case A and E. For Case D, which has less liquid cloud amount overall than Case A and E, the variations of LWP are strongly influenced by the radiative effect of light absorbing aerosols. (Figure 9). The heating in the free troposphere

induced by aerosols can reduce the LWP of the stratus by 25% in the polluted scenario during the Case D1, and even reverse the trend of LWP responses to aerosols from positive to negative. Similarly, for the cumulus in D2, LWP is reduced by about 20% comparing DIE and IEO for both moderate and heavy polluted aerosol conditions.

3.5 Radiative Fluxes and Surface Temperature

In all three cases, the modeled shortwave radiation reaching the surface shows a monotonic decrease by increasing aerosols, while the magnitude of the decrease depends on cloud type and is affected by the aerosol radiative effect. For the IEO cases, the general reduction in shortwave radiation agrees with the monotonic increase in LWP but decrease in cloud droplet radius. By considering the aerosol direct effect, Case A and D show much larger reduction of surface shortwave radiation, as large as -30 W m^{-2} (Figure 10). In contrast, due to the thick cloud layer in Case E, the aerosol radiative effect on the surface radiation is not evident for both clean and polluted condition. On the other hand, we find that the cooling from the aerosol indirect effect is most significant in Case E which has larger cloud thickness and more persistent low-level stratiform cloud than the Case A and D. Comparing the aerosol-induced trends of SW radiation in IEO runs among three cases, Case E exhibits the largest reduction of SW radiation by increasing CCN.

The response of outgoing longwave radiation (OLR) at the top of the atmosphere to aerosol variations is modulated by overall cloud fractions. Such a hypothesis can be verified by comparing the responses of OLR (Fig. 10) and cloud fraction (Fig. 8). It is found that almost all the trends of OLR are opposite to those in cloud fraction, as OLR (cloud fraction) decrease (increase) in DIE of Case A, IEO of Case D, both DIE and IEO

of Case E, while increase (decrease) in IEO of Case A and DIE of Case D1. A quantitative analysis of the co-variations of CF and OLR in Fig. 11 shows the correlation coefficient (R) is greater than 0.8 between those two quantities. This good agreement reinforces the fact that clouds play a crucial role in altering the earth radiative budget regionally and globally. Comparing DIE and IEO cases, the aerosol radiative effect can even change the sign of the OLR response. For example, in Case A, introducing aerosol-radiation interactions results in a change of OLR due to aerosols from $+2.4$ to -2.4 W m^{-2} . Even though cloud top height is another potential factor to alter OLR, we find the cloud top heights do not significantly change for different aerosol conditions in all three cases, as evidenced by the vertical profiles of cloud fraction (Fig. 8) and ice water content (Fig. S3)

Surface temperature changes closely follow surface radiation imbalance. The model simulated surface temperature shows good agreement with surface station measurements at SGP. In Case A and D, only aerosol radiative effects in the polluted scenario stand out and produce a surface cooling (Figure 12). The insignificant surface temperature response in IEO can be attributed to the cancellation between both enhanced shortwave cooling and longwave warming at the surface. In contrast, the surface cooling induced by aerosols is significant in both IEO and DIE in Case E. Overall, the responses of surface temperature to aerosol variations exhibit large similarity with those of downwelling shortwave radiative fluxes at the surface (Figure 10).

4. Conclusion

The individual mechanisms of aerosol-cloud interaction in certain type of cloud have been extensively examined by previous studies using similar cloud-resolving

models. However, aerosol effects on cloud ensembles where multiple types of clouds coexist in the real atmosphere receive much less attention so far. In this study we adopted an “ensemble” approach and compiled three cases that were well observed over the same region (US SGP) during the same season (springtime). An aerosol-aware WRF model is used to explore the differences in the responses of cloud micro- and macro-physics, precipitation, and radiation to aerosol perturbations in the complex continental cloud systems. The model employs a two-moment bulk microphysics scheme to account for the aerosol microphysical effect, as well as a modified Goddard radiation scheme to simulate the aerosol radiative effect. Three different cloud systems during the March 2000 Cloud IOP campaign at the ARM SGP site were examined, including: a low pressure system, less-precipitating stratus and shallow cumulus, and a cold front. The impacts of increasing aerosol concentration are quantified along with the comparison between aerosol radiative (direct and semi-direct) and microphysical (indirect) effects. Our cloud-resolving simulations generally captured the major features of observed temporal variations of precipitations for all the three cases.

The sensitivity experiments show that the distinct microphysical responses of cloud collections to initial aerosol loadings highly depend on the cloud types and synoptic conditions. For the convective cases with moderate-to-heavy precipitation, the microphysical properties for hydrometeors show robust monotonic trends in response to aerosol loadings, evident in the dependences of the mass content and number concentration of cloud droplets and rain drops on aerosols. The inclusion of aerosol direct effects in those cases has little influence on the monotonicity of the microphysical response for the heavy precipitating situation. For the less-precipitating stratus clouds, the

monotonicity of the microphysical response to aerosols depends on the types of hydrometeors examined, as the cloud water content can be strongly modulated by aerosol direct and semi-effects.

The overall response of domain-averaged accumulated precipitation to aerosol initial concentrations shows a linearly increasing trend for convective clouds but some tipping points in stratiform or shallow clouds. The monotonicity of the precipitation response to aerosol initial concentrations differs from case to case and varied with cloud types and their evolving stages, indicating a high dependence of precipitation response on the weather environment in which cloud systems developed. By comparing the IEO simulations with DIE simulations, the importance of aerosol direct effects emerges to determine the sensitivity of precipitation to aerosol, although the aerosol direct effects do not modify the trends of precipitation in response to aerosol perturbations.

The changes in cloud macrophysics like cloud fraction do not show any similarity with those in cloud microphysics. The responses of cloud fraction to different aerosol concentrations are quite distinctive in different cases. Specifically, in a convective cloud system like case E (Figure 8d), we found the increase in cloud fractions is due to the increase in liquid cloud mass in the stratiform clouds trailing the deep convection core. In a less-precipitating cloud system like our case D, the absorbing aerosols can heat up the air mass in the cloud layer, cool the air near the surface, reduce the RH in the cloud layer, weakened the turbulence in the boundary layer, and eventually decrease cloud fraction (Figure 8b,c). Note that the reports of the aerosol effects on cloud fraction are mainly from cloud-resolving simulations (e.g. Lin et al., 2016), while GCMs always predict little changes of cloud fraction in response to aerosols perturbations (e.g. Wang et al., 2015). It

stresses the importance of cloud fraction parameterizations in GCMs. LWP exhibits a robust monotonic increase along with the elevation of aerosol loading for convective clouds and their trailing stratiform clouds, while there is no significant change in LWP for the thin mid-level cloud with little water content inside. The modeled shortwave radiation reaching the surface shows a monotonic decrease by increasing aerosols, while the magnitude of the decrease depends on cloud type and is affected by the aerosol radiative effect. The responses of OLR are closely linked with the total cloud fraction under different aerosol loadings. In the end, the surface temperature changes closely follow surface radiation imbalance including both shortwave and longwave contributions, while the former carries more weight.

The results from this modeling study highlight the complexity of the aerosol-cloud-precipitation-radiation interactions that vary on a case-by-case basis. In addition, this study has shown that studying the aerosol microphysical effect alone is not sufficient to assess the changes of the clouds in the real atmosphere, as the aerosol radiative effects can also produce profound impacts on cloud development and precipitation processes. Therefore, long-term high-resolution model simulations with comprehensive aerosol effects are greatly needed to quantify the climatic effects of aerosols on the regional radiation budget and the hydrological cycle.

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Figures

Figure 1. a) The three nested domains used in the WRF model in this study. The innermost domain has 2 km spatial resolution. The diamond represents the ARM SGP Central Facility. b) The three vertical aerosol profiles used in this study: clean (green), SGP (yellow), and polluted (red).

Figure 2. Surface pressure and wind maps (left) and satellite images (right) for each of the three cases in this study: a) 0300Z on March 3, 2000 (low-pressure system); b) 1200Z on March 14, 2000 (non-precipitating stratiform); and c) 0300Z on March 16, 2000 (cold front). Satellite images were taken 15 minutes before the listed times.

Figure 3. Domain averaged water mixing ratios of cloud, rain, and ice hydrometeors for Case A, Case D1 (stratus period), case D2 (cumulus period) and Case E, with 1, 2, and 3 coinciding with clean, SGP, and polluted aerosol profiles. The black lines indicate the direct-and-indirect-aerosol-effect (DIE) cases, while the blue lines indicate the aerosol-indirect-effect-only (IEO) cases..

Figure 4. Domain averaged number concentration of cloud, rain, and ice hydrometeors for Case A, Case D1, Case D2 and Case E, with 1, 2, and 3 coinciding with clean, SGP, and polluted aerosol profiles and the black and blue lines coinciding with DIE and IEO model runs, respectively.

Figure 5. The vertical-temporal profiles of domain-averaged air temperature changes due to aerosol radiative effect in three aerosol concentrations for Case A. The times shown in the plot are UTC.

Figure 6. The vertical-temporal profiles of changes in effective radius of cloud droplet, raindrop, and ice crystal between the clean and pollute conditions for Case A, Case D and Case E. Only IEO results are shown here.

Figure 7. Temporal evolution of observed and simulated rates (left column panels) and accumulated precipitation (right column panels) for Case A, Case D and Case E. Plotted colors correspond as follows: black—observed, green—clean DIE, yellow—SGP DIE, red—polluted DIE, cyan—clean IEO, blue—SGP IEO, dark red—polluted IEO. For right column panels, with 1, 2, and 3 coinciding with clean, SGP, and polluted aerosol profiles and the black and blue lines coinciding with DIE and IEO model runs, respectively. Significant for 95% confidence intervals.

Figure 8. Simulated and observed cloud fraction for Case A (a,e,f,g), Case D (b,c,h,i,k), and Case E (d,l,m,n). First row: averaged cloud fractions over the entire time period. Number 1, 2, and 3 denote clean, SGP, and polluted aerosol profiles, respectively; the second to fourth rows: clean (e, h, l) and polluted (f, i, m) DIE simulations, as well as observations (g, k, n).

Figure 9. Simulated and observed liquid water path (LWP) for Case A (e), Case D (f), and Case E (g). First row: averaged over the entire time period and the innermost domain. Number 1, 2, and 3 denote clean, SGP, and polluted aerosol profiles, respectively; Second row: LWP are averaged for the entire domain. Plotted colors correspond as follows: black—observed, green—clean DIE, yellow—SGP DIE, red—polluted DIE, cyan—clean IEO, blue—SGP IEO, dark red—polluted IEO, and for (c) lavender—observations.

Figure 10. Simulated surface downwelling shortwave radiation fluxes (first row) and TOA outgoing longwave radiation (OLR, second row) for Case A (first column), Case D (second and third columns), and Case E (fourth column). Number 1, 2, and 3 denote clean, SGP, and polluted aerosol profiles, respectively.

Figure 11. Correlation between total cloud fraction and OLR from three cases under different aerosol conditions.

Figure 12. Simulated and observed surface temperature for Case A (e), Case D (f), and Case E (g). The first row shows averaged surface temperature over the entire time period and the whole domain. The second row shows the temperature evolution in three cases. Plotted colors correspond as follows: black—observed, green—clean DIE, yellow—SGP DIE, red—polluted DIE, cyan—clean IEO, blue—SGP IEO, dark red—polluted IEO, and for (c) lavender—METAR temperature observations.

Support Figures

Figure S1. The vertical-temporal profiles of domain-averaged core vertical velocity under different aerosol pollution conditions for Case A.

Figure S2. The vertical-temporal profiles of domain-averaged core vertical velocity under different aerosol pollution conditions for Case D.

Figure S3. Case E domain averaged ice water mixing ratio for (a), (b), and (c) clean, SGP, and polluted DIE cases, respectively; (d), (e), and (f) clean, SGP, and polluted IEO cases, respectively.

Figure S4. The temporal profiles of temperature perturbations due to aerosol direct effects (DIE-IEO) for case A.