1 Improving aerosol interaction with clouds and precipitation in a regional

2	chemical weather modeling system
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# 7 Abstract

A comprehensive aerosol-cloud-precipitation interaction (ACI) scheme has been 8 developed under CMA chemical weather modeling system GRAPES/CUACE. 9 Calculated by a sectional aerosol activation scheme based on the information of 10 size and mass from CUACE and the thermal-dynamic and humid states from the 11 weather model GRAPES at each time step, the cloud condensation nuclei (CCN) is 12 fed online interactively into a two-moment cloud scheme (WDM6) and a 13 convective parameterization to drive the cloud physics and precipitation 14 formation processes. The modeling system has been applied to study the ACI for 15 January 2013 when several persistent haze-fog events and eight precipitation 16 events occurred. 17

The results show that interactive aerosols with the WDM6 in GRAPES/CUACE obviously increase the total cloud water, liquid water content and cloud droplet number concentrations while decrease the mean diameter of cloud droplets with varying magnitudes of the changes in each case and region. These interactive micro-physical properties of clouds improve the calculation of their collection growth rates in some regions and hence the precipitation rate and distributions in

24	the model, showing 24% to 48% enhancements of threat score for 6-h
25	precipitation in almost all regions. The interactive aerosols with the WDM6 also
26	reduce the regional mean bias of temperature by 3 °C during certain precipitation
27	events, but the monthly means bias is only reduced by about 0.3°C.
28	

29

## Introduction

Aerosols can act as cloud condensation nuclei (CCN) or ice nuclei (IN) to 30 participate in cloud formations, alter the microphysics and life time of clouds and 31 then impact the precipitations(Twomey, 1977;Ramanathan et al., 2001;Seinfeld 32 and Pandis, 1997; Albrecht, 1989), which is so-called aerosol cloud interactions 33 (ACI). Most of the previous researches on ACI focus on single clouds or climate 34 impacts with very limited numbers on the meso-scale weather, partially due to the 35 difficulties to establish real and reasonable connections from emissions to aerosol, 36 CCN, clouds and then precipitations. Aerosols, impacted by emission, processes of 37 microphysics and atmospheric thermo-dynamics, are in different size ranges and 38 consisted of several components which are often temporally and spatially varied 39 (Jacobson et al., 1994;Zhang et al., 1999;Zhang et al., 2012a;Gong et al., 2003). 40 Some components, such as organic carbon (OC), are in very complex structures 41 and many of their precursors still cannot be detected (Stockwell et al., 1997;Fuzzi 42 et al., 2006; Jacobson et al., 2000). Meanwhile clouds are also the results of 43 complex interactions among atmospheric thermo-dynamics of different processes. 44 The scale of aerosol-cloud interactions spans from nanometer to thousand 45 kilometers including both complex microphysics and scales of clouds and aerosols 46 and macrophysics of air mass and atmospheric circulations. All of these make it 47 very difficult to establish a direct connection from emission to precipitation to 48 quantify the effects of aerosol on clouds and precipitations in both climate and 49 weather models (Khain, 2009;Lohmann and Feichter, 2005;Stevens and Feingold, 50

51 2009;Tao et al., 2012).

A weather model with aerosol feedbacks is an important and effective way to 52 explore the interactions between aerosols, clouds and precipitation (Yin et al., 53 2002;Levin and Cotton, 2009;Tao et al., 2012;Khain, 2009). Currently, two distinct 54 approaches, the bin method and the bulk method, are often used to explore cloud 55 microphysics in the weather models. For bin models, each type of cloud 56 hydrometers is sectionally resolved to represent their mass or size distributions 57 which might be changed in the course of the model integration (Khain and Sednew, 58 1996; Khain et al., 2004). The bin methods can explicitly resolve cloud microphysics 59 and provide very rigorous solution than bulk approach, but they are limited to 60 single clouds because of huge computation. The bulk models have been improved 61 from single-moment approach to two-moment approach. The two-moment 62 models can predict not only mass but number concentrations which allow more 63 flexibility of the size distribution and enable the mean diameter to evolve in 64 contrast with the single-moment method (Morrison et al., 2005;Seifert and 65 Beheng, 2001;Lim and Hong, 2010). 66

<sup>67</sup> However, no matter which cloud approach is used, the relationship between <sup>68</sup> aerosol and cloud droplet needs to be established for ACI. One of the existing <sup>69</sup> relationships is the Twomey approach which links the cloud droplet number <sup>70</sup> concentration to aerosols number concentration ( $N_0$ ) by two simplified <sup>71</sup> parameters, the supersaturation (S) and its power exponent ( $\kappa$ ), and has been <sup>72</sup> widely used in climate and weather models (Ramanathan et al., 2001;Gultepe and

Isaac, 1996; Boucher, 1995; Twomey, 1959; Khairoutdinov and Kogan, 2000). In this 73 approach, the two parameters  $N_0$  and  $\kappa$  should vary from regions to regions. While 74  $\kappa$  has been set to be constant and N<sub>0</sub> has been roughly classified into four zones: 75 rural, urban, ocean and remote continents with no connections to the emissions 76 and production of the actual aerosol distribution in most cases and with no 77 physical processes of aerosol activations, i.e. CCN. Even though in some models 78 CCN can be prognostic as a result of advection, activation and scavenging, aerosol 79 size distribution and the total aerosol number have to be prescribed (Khain et al., 80 2004; Fan et al., 2012; Yin et al., 2002). This may introduce additional bias into 81 aerosol and cloud interaction, especially for regions like China with high aerosol 82 concentration and diverse compositions. 83

East China is one of the most polluted areas in the world (Zhang et al., 2012a). 84 High accumulated aerosols and stagnant weather systems frequently help to 85 form the regional haze-fog evens there (Horton et al., 2014; Zhang et al., 86 2013a;Zhang et al., 2013b;Che et al., 2014).Heavy aerosol pollution has been 87 related to the decrease precipitation and the cooling radiative effect elsewhere 88 (Cheng et al., 2005;Zhao et al., 2006;Ma et al., 2010;Wang et al., 2015). Several 89 studies by two-moment scheme with the highly parameterized scheme Twomey 90 formula or by bin model for one cloud show that microphysics of different regimes 91 of clouds and precipitation can be more realistically simulated by adding aerosol 92 impacts in China (Zhang et al., 2007;Guo et al., 2014;Fan et al., 2012;Yang et al., 93 2011). Since high aerosol concentrations alter the radiation, cloud microphysics 94

and then the precipitation, its impacts on weather systems cannot be ignored in
regional weather models.

In order to take into account of the effects of aerosol on cloud and precipitation, 97 a comprehensive scheme containing the emission, aerosols, clouds and their 98 interaction mechanisms has been developed in GRAPES/CUACE. It is built on the 99 base of the China Meteorological Administration (CMA) Unified Atmospheric 100 Chemistry Environment/Aerosol (CUACE/Aero)(Zhou et al., 2012), and the CMA 101 weather forecasting model Global/Regional Assimilation and PrEdiction System 102 (GRAPES) (Chen et al., 2008). By integrating a time dependent CCN formulation 103 from an aerosol activation scheme directly from CUACE, it enables the 104 quantitative assessment of the impacts of aerosol pollution on clouds and 105 precipitation. 106

By developing and using the ACI scheme in GRAPES/CUACE here, the aerosol 107 impacts on clouds and precipitations in East China have been investigated for 108 January 1-31, 2013 when a series of long lasting haze-fog episodes hit this region 109 and eight precipitation events occurred. This paper has divided into five sections, 110 beginning with Section 2 for the description of the modeling system: 111 GRAPES/CUACE. Case description and numerical experiments designs are in 112 Section3. Results and discussions will be shown in Section4, followed by Section 5: 113 Conclusions. 114

#### **115 1. ACI scheme established in GRAPES/CUACE**

#### 116 2.1 GRAPES/CUACE System

The CMA new generation of weather forecasting model GRAPES is a fully 117 compressible non-hydrostatical weather model systems by using a semi-implicit 118 and semi-Lagrangian discretization scheme (Zhang, 2008;Xu et al., 2008;Chen et 119 al., 2008). It uses a Arakawa C staggered grid and the central finite-difference 120 approach of second-order accuracy in horizontal coordination, a nonhydrostatic 121 approximation method together with the staggered approach of Charney-Phillips 122 to improve the accuracy of vertical pressure gradients and a semi-implicit and 123 semi-Lagrangian scheme for temporal and advection discretion. A height-based 124 terrain following coordinate, which behaves as a natural height coordinate, is used 125 to compromise the Lagrangian trajectory errors in spherical coordinates at high 126 latitudes. The physical packages include cumulus convective, single moment cloud 127 microphysics, radiative, land surface and boundary layer processes. 128

CUACE is a unified atmospheric chemistry environment with four major 129 functional sub-systems: emissions, gas phase chemistry, aerosol microphysics and 130 data assimilation. It is designed to facilitate the establishment of a chemical 131 weather forecasting system using near real time data in China(Zhou et al., 132 2012; Zhou et al., 2008). Seven aerosol components, i.e. sea salts, sand/dust, 133 elemental carbon, organic carbon, sulfates, nitrates and ammonium salts are 134 sectioned in 12 bins with detailed microphysics of hygroscopic growth, nucleation, 135 coagulation, condensation, dry depositions and wet scavenging in the aerosol 136

module. The gas chemistry module is based on the second generation of Regional

Acid Deposition Model (RADM II) mechanism with 63 gaseous species through 21

photo-chemical reactions and 121 gas phase reactions applicable under a wide

variety of environmental conditions especially for smog(Stockwell et al., 1990) and

141 prepares the production rates of sulfate and secondary organic aerosol for the

aerosol module. Emission inputs have been provided by the same emission

<sup>143</sup> subsystem EMIS of CUACE with the official basic emission sources data updated to

144 the year of 2010 (Zhou et al., 2012).

145 CUACE is fully online coupled to the regional version of GRAPES to establish the

comprehensive modeling system – GRAPES/CUACE and has been used for

radiative feedback researches (Wang et al., 2010; Wang et al., 2015).

148 2.2 Aerosol activation Scheme HG

149 The sectional aerosol activated scheme developed by Hayder Abdul-Razzak

and Ghan (HG scheme) provides a convenient platform to connect sectional

aerosols into cloud physics(Abdul-Razzak and Ghan, 2002;Hayder and Ghan,

152 2000;Abdul-Razzak et al., 1998;Ghan et al., 1995;Ghan et al., 1993). This scheme

is derived from the basic theory of Köhler curve to calculate how a particle can

be activated under a certain supersaturation in an air parcel. The newly activated

155 CCN would be parameterized in terms of environmental supersaturation and the

upper and low limit of the critical supersaturation for each aerosol bin. This

157 method is very useful for precise determination of concentration of droplets

nucleated at the cloud base. However, droplet nucleation or aerosol activation

can also take place above the cloud base as well, which is induced by increasing 159 supersaturation above cloud base and lateral entrainment of the surrounding air 160 with dry aerosols (Khain et al., 2000). Therefore, it is not enough to calculate the 161 CCN at the cloud base only in an air parcel, especially for stratus clouds where 162 turbulence is more important than the vertical movement (Bodenschatz et al., 163 2010). For these reasons, the HG scheme has been online coupled with both the 164 stratus scheme and the convective scheme in GRAPES. Consequently, Aerosols 165 from CUACE can be activated into CCN as the humid condition is satisfied, not 166 just in the cloud base. 167 2.3 Aerosol activation in Warm Double-Moment 6 scheme 168 The WRF Double-Moment 6-class scheme(WDM6) is introduced into GRAPES 169

since it can predict not only the mass but also the number of droplets and drops
(Lim and Hong, 2010). It is developed from the WRF Single-Moment 6-class
scheme (WSM6) and needs the CCN input for cloud droplets which provides a
direct way for aerosol feedbacks into clouds and precipitation. The original
activation scheme is expressed as (K2000) (Twomey, 1959;Khairoutdinov and
Kogan, 2000):

176 
$$n_a = \left(n + N_c\right) \left(\frac{S_w}{S_{\max}}\right)^k$$
(1)

Where n is the aerosol number concentration, κ is the activation power exponent
 S<sub>max</sub> is the max supersaturation to activate all the aerosols, S<sub>w</sub> is the air
 supersaturation. The change rate of cloud droplet number concentration due to
 CCN activation is given by:

$$\frac{\partial N_c}{\partial t} = \frac{\max\{0, (n+N_c)\min\{1, \left(\frac{S_w}{S_{max}}\right)^{\kappa}\} - N_c\}}{\Delta t}$$
(2)

181

In this activation scheme: n, κ and max S<sub>max</sub> are usually preset just like the way Twomey formula does with the inherited shortcomings due to the insufficient information of real aerosols. To overcome these, the aerosol size and composition information from CUACE and the humid and thermal information from GRAPES are used in HG scheme to calculate the CCN at each time step, which is then fed into WDM6 to replace the preset CCN by the scheme of K2000.

An aerosol particle can be quickly activated into CCN, usually in less than a 188 second, as the atmosphere reaches the critical supersaturation of the aerosols 189 (Kogan, 1991). In models with relatively coarse resolution like GRAPES, 190 supersaturation cannot be easily satisfied because of the insufficient information 191 of inhomogeneous turbulence and vertical movements even when the grid-mean 192 relative humidity is over about 85 to 90%. While local supersatureation around a 193 particle can be satisfied through which clouds can be formed. As the local 194 supersatureation is not only decided by the mean fields, a parameterized scheme 195 has been developed to add the effect of local turbulence and vertical movements 196 to the supersaturation in HG scheme for WDM6 in GRAPES. The turbulence effect 197 is assumed to be proportional to the mean relative humidity and inversely 198 proportional to both the horizontal wind speed and the vertical height. 199

200 2.4 Aerosol activation in KF scheme

Stratiform and convective precipitations are the two precipitation regimes that 201 cannot be mutually excluded. The former refers to those precipitations with low 202 vertical motions and the later with stronger ones(Houze, 1997). These two 203 regimes can be fully explicit in meso-scale model only under the condition that 204 the model resolution is below several hundred meters(Molinari and Dudek, 205 1992), which is very difficult to realize for the on-line coupled modeling systems 206 such as GRAPES/CUACE. A hybrid approach is often used in most meso-scale 207 models which can explicitly condense water for the stratus precipitation in grid 208 scale, to parameterize convective scale precipitation in sub-grid. In order to fully 209 account the ACI in the model, HG scheme has also been coupled to the Kain-Fritch 210 211 convective parameterization scheme (KF) in GRAPES/CUACE (Kain, 2003;Kain and Fritsch, 1990; Fritsch and Chappell, 1980). The sectional aerosols have been 212 introduced and moved upward or downward as the air mass does in KF. They 213 would be activated as soon as the supersaturation satisfies the critical 214 supersaturation by HG scheme. 215

Since there is no size information for cloud and rain water in the bulk
convective scheme, a generalized gamma distribution has been introduced to
describe the cloud-droplet and raindrop spectra (Cohard and Pinty, 2000;Walko et
al., 1995;Clark, 1974).

220 
$$N_X(D_X) = N_X \frac{\alpha_X}{\Gamma(\nu_X)} \lambda_X^{\alpha_X \nu_X} D_X^{\alpha_X \nu_X - 1} \exp\left[-\left(\lambda_X D_X\right)^{\alpha_X}\right]$$
(3)

Here 
$$\lambda_x$$
 is the slope parameter,  $\alpha_x$  and  $v_x$  are two shape  
parameters.  $\lambda_x = \left[\frac{\pi}{6}\rho_w \frac{\Gamma(v_x + 3/\alpha_x)}{\Gamma(v_x)} \frac{N_x}{\rho_a r_x}\right]$ ,  $N_x = N_0 + N_{cen}$ ,  $N_{cen}$  is the newly  
activated aerosol number concentration,  $N_0$  is the cloud number concentration  
which is set to be 30 cm<sup>-3</sup> in terms of observations at autumn in east China(Zhang  
et al., 2012b;Zhang et al., 2011). Normally  $\alpha_x$  can be specified priori as 1 for  
number concentration size distribution and 3 for mass concentration size  
distribution. Only  $v_x$  left has to be tuned through measurements. A  
parameterization for  $v_x$  has been proposed through the total droplet number  
concentration and liquid water content(Geoffroy et al., 2010):

230 For size spectra of number concentration:

231 
$$\alpha_x = 1, \quad v_x = 14.5q_c + 6.7$$
 (4)

For size spectra of mass concentration:

233 
$$\alpha_X = 3, \quad \nu_X = 1.58q_c + 0.72$$
 (5)

Here  $q_c$  is the liquid water content in the unit of g m<sup>-3</sup>. With Eqs. (4) and (5), droplet size distribution in (3) can be bulky represented by the total liquid water content and total cloud droplet number concentration which can then include the newly activated aerosols from HG.

The condense of KF scheme is allowed to be removed as precipitation based on an empirical relationship(Ogura and Cho, 1973).

240 
$$\delta r_c = r_{co} (1 - e^{-c_1 \delta z / w})$$
 (6)

Here  $c_1$  is a constant,  $\delta_2$  is the height of the layer. The precipitable water  $\delta_{r_a}$ 241 in (6) is inversely proportional to the vertical velocity w, which means that a 242 small vertical velocity would produce more  $\delta r_c$ . As smaller vertical velocity means 243 weaker convection and less  $r_{ca}$  (total condense), less precipitable water  $\delta r_c$  would 244 be produced. Therefore,  $r_{co}$  and w are offset each other, which may cause false 245 large convective precipitation. In order to overcome this problem, the size 246 spectrum information of cloud drops and droplet are introduced into the 247 convective clouds, which resolves the total cloud water content and number 248 concentrations including the newly formed CCN. The new precipitable rain water, 249 named as  $r_p$ , can be calculated by integrating (3) to replace  $r_{co}$  in (6). The 250 threshold for cutoff radius between the droplets and rain drops in the integration 251 is 50 µm for deep convective precipitation (Berry and Reinhardt, 1974; Seifert and 252 Beheng, 2001), and 25 µm for shallow convective precipitation (Khairoutdinov and 253 Kogan, 2000). Terminal velocity  $w_T$  for the rain drops has also been introduced. 254 Finally, Equation (6) is reformed as: 255

256

$$\delta r_c = r_p (1 - e^{-w_T / w}) \tag{7}$$

Now, the comprehensive interactions of aerosol-cloud-precipitation have been
 established in GRAPES/CUACE with activated aerosols from CUACE linked into
 both the stratus and convective clouds to participate in the cloud and
 precipitation processes.

## 261 **2. Case description and numerical experiment designs**

#### 262 3.1 Case description

A series of long-lasting heavy haze-fog events hit east part of China in January, 263 2013. Climatology analysis shows that the mean number of the hazy days in this 264 period is much higher than the mean value from 1981 to 2010, especially in the 265 three major pollution zones of North China Plain, Delta of Yangtze River and Zhu 266 River(Zhang et al., 2013a). Meanwhile, the values of the stagnant polluted 267 parameter PLAM, a threshold value to distinguish clear and polluted weather, are 268 over 80 in most part of east China, which indicates strong static weather 269 conditions for pollutant accumulation (Zhang et al., 2013b; Wang et al., 2012). 270 Surface daily mean  $PM_{2.5}$  concentrations are in the range of 100-150 µg m<sup>-3</sup> and 271 AOD is above 1.0 in many surface stations (Che et al., 2014; Wang et al., 2014). 272 Eight precipitation events also occur in January 2013 with six of them 273 sweeping over regions south of Yangzi River as every low pressure system moving 274 out of Sichuan Basin (Table 1). Other two cases are related to the cold fronts and 275 affect the whole east of China. This forms a very good period to study ACI under 276 high pollution conditions. 277 3.2 Numerical experiment designs 278 Three sets of experiential runs are designed: T1 with the single-moment 279 microphysics scheme of WSM6 and original KF without aerosol activation; T2 with 280 WDM6 scheme and the activation scheme K2000 and T3 with HG activation 281

scheme online connected with WDM6, KF and CUACE. Five target regions are

selected for the evaluation in terms of typical heavy pollution regions over China

(Fig. 1). R1 and R2 are covering the whole China and East China respectively.

285 Within R2, R3, R4 and R5 are representative of three typical polluted zones: North

- East China, North China Plain and South China.
- The meteorological initial and boundary conditions, at the resolution of 0.5°,

are interpolated from the forecasting outputs of the CMA medium Meteorological

model T639 in 6-hour interval. The surface daily and hourly PM<sub>2.5</sub> concentrations

<sup>290</sup> from CMA Atmosphere Watch Network (CAWNET) are used to evaluate the

<sup>291</sup> performance of aerosols (Wang et al., 2008;Zhang et al., 2012a). Precipitation

data from the rain gauges in meteorological stations over China are used for the

<sup>293</sup> precipitation threat scoring (Wang et al., 2008). Temperature, geopotential height,

<sup>294</sup> humidity and cloud water mixing ratio of NCEP reanalysis at standard pressure

- levels from 1000 hPa to 10 hPa are used to evaluate the outputs of
- 296 GRAPES/CUACE (Kalnay et al., 1996).

#### **3. Results and Discussions**

In order to quantify the impacts of aerosols on precipitations, the cloud
properties such as the total cloud water content, clouds liquid water content and
mean droplet diameters are analyzed to elucidate the aerosol's effect on clouds.
As hourly changes of thess variables above are very chaotic, regional means are
discussed in order to avoid the interruption of small scale advection and diffusion.
The threat scores (TS) of 6-h precipitation are also quantitatively analyzed to
evaluate the aerosol's effect on precipitation. The changes in temperature and

<sup>305</sup> height are also discussed to explore the aerosol effects on the thermal and

306 dynamics. Hourly surface aerosol concentrations are compared to the

307 measurements to see the ACI feedbacks on aerosol distribution.

308 4.1 Aerosol Effects on the Clouds

Regional-monthly-mean vertical profiles of the total cloud water (the total mass 309 of cloud water, rain water, ice water, snow and grauple) and temperature for T1, 310 T2 and T3 together with the NCEP reanalysis in R1, R2, R3, R4 and R5 are shown in 311 Figure 2 a1-a5 and b1-b5. Compared to T1, T2 and NCEP reanalysis in the layers 312 below 600 hPa, the total cloud water all increases obviously in the five regions for 313 T3 with a clear peak at about 850 hPa. These results indicate that with more 314 realistic aerosols interacting with clouds (T3), more water vapor condenses into 315 cloud water due to the activation of aerosol particles. 316

The amount of the increasing cloud water for T3 has very unique regional 317 characteristics. The increase in R1, i.e. national wide with plenty of remote dry 318 areas such as Tibetan and Northwest China, is much less than in R2 which 319 represents the most developed regions in China with a lot of emissions. Of the 320 three typical polluted regions, R3 covers most part of Northeast China with 321 below-freezing temperature, which is not favorable for warm cloud formation and 322 therefore no obvious increases is found compared between T2 and T3. While the 323 temperature in R4, the most polluted area in China, is near or just above freezing, 324 the condition is favorable for the long lasting haze formation but not a good 325 condition for cloud and precipitation formation with the relatively less cloud water. 326

In R5 where abundant cloud water exists and temperature under 700 hPa is above
 freezing, this region has the most enhancement for aerosol activation and warm
 clouds development.

The amount of total cloud water by T1 and T2 is compatible to each other 330 because there is no aerosol feeding into the single moment scheme T1 and the 331 aerosol activation is based on the prescribed aerosol numbers without spatial 332 variation in T2. Profiles of cloud water from NCEP are close to that of T1 and T2 in 333 all of the five regions. Even though the basic variables such as wind, temperature, 334 relative humidity, height and pressure are most analyzed from measurements, the 335 cloud water from NCEP reanalysis is not from direct observations but from cloud 336 physics diagnosis. Cloud water data from NCEP reanalysis should be carefully used 337 in the regions affected by high aerosol concentrations. 338

For all regions, the increases by T3 in total cloud water contents are driven by liquid water contents (Fig. 3 a1-f1). The solid cloud water for T1, T2 and T3 is in the same order of 1.0E-3 g kg-1 in all the three regions and follow almost the same vertical distribution shapes with tiny difference of height and thickness in each region, showing relatively small effects from aerosols ((Fig. 3 a2-f2).

The aerosols activation in T3 can also increase the cloud number concentrations which affect the mean diameters of droplet (MDD). The profiles of

MDD in R4 and R5 are shown in Figure 4. MDDs for T3 are all below 10 µm, about

one order less than those of T2, and change little with height in the lower

troposphere under 700 hPa (Fig. 4a), showing a clearly decrease after aerosols

activated into clouds in North China Plain (R4) and South China (R5).

Conventionally, smaller droplets (usually less than 20 µm) need more time to grow
up into rain drops than the larger ones, which should result in more rains
produced in T2 than in T3 after a decrease in MDDs. However, due to the
self-collection growth process, which depends on both the MDDs and number
concentrations of cloud droplets, the simulations in T2 do not see more rain
produced (as discussed in Section 4.2).

In order to investigate the combined impacts of both MDDs and number 356 concentrations on rain production, the self-collection kernels for the 357 two-moments WDM6 from Long's work (Long, 1974) is used. As showed in Fig. 4e, 358 self-collection rates (SCRs) of R5 for T3 are the largest among all the three tests 359 among the three regions of R3, R4 and R5. They are over 100 m<sup>-3</sup> s<sup>-1</sup> under 700 hPa, 360 about one order of magnitude higher than by T2. This indicates that in R5 even 361 though the activated aerosols from CUACE decrease the MDD which may in some 362 way decrease the self-collection process, the high concentration of cloud droplets 363 from aerosols activation, four orders of magnitudes higher than that of T2, 364 enhances the chance of collision and compromises the decreasing collision trend 365 by MDD. Therefore, the SCRs can explain the phenomenon that the precipitation 366 simulation ability increases obviously for T3 in R5 (in Section 4.2). While in North 367 China Plain (R4), SCRs for T3 and T2 are almost the same, varied around the value 368 of 2 m<sup>-3</sup> s<sup>-1</sup>, meaning that a lot of aerosols activate as a small cloud drops , grow 369 very slowly into larger ones and stay relatively longer in the atmosphere, forming 370

the long lasting haze there (Fig.4d). In Northeast China (R3), SCRs for T3 and T2
are so small that the SCRs could be ignored, which is also consistent with the cold
cloud formation (Fig. 4c).

In summary, the aerosol effects on cloud formation are very different in the 374 three typical polluted regions: R3, R4 and R5. As shown above, R3 is controlled by 375 cold cloud formation processes, little effect can be seen in this region. In R4, the 376 relatively humid layer from the surface to 600 hPa indicates a favorable condition 377 for the long lasting haze formation as proved by both the mean diameter of 378 droplets and collection rates. In R5, liquid water for T3 is also one order higher 379 than that in R4 and three orders of magnitude higher than that in R3. Together 380 with the highest collection rate, R5 processes a good condition for precipitation 381 formation, which is consistent with the improvement of the precipitation 382

simulation ability as discussed below.

4.2 Aerosol effects on precipitation

385 4.2.1 Regional Threat Scoring Evaluation

The threat scoring is a common and useful way to quantitatively evaluate the 386 model performance of regional precipitation (Mitternaier et al., 2013; Gilleland et 387 al., 2009). The 6-h accumulated gauge values from 1400 routine weather stations 388 in CMA are used for the evaluation. The threshold value for the contingency table 389 is 0.1 cm which is in harmony to the 24-h's threat scoring threshold usually used in 390 most operational weather forecasting centers. Threat scoring for 6-h precipitation 391 is stricter than 24-h's due to the short time scale. The model precipitation results 392 from the three tests T1, T2 and T3 are interpolated into the meteorological 393

stations and compared with the observations through the 2 X 2 contingency table 394 to calculate the threat scores for the five targeting regions: R1, R2, R3, R4 and R5. 395 The time series of 6-h precipitation threat score for T3 are consistently higher 396 than those for T2 and T1 for all five regions (Figure 5a-5d) with a monthly-mean 397 improvement of about 33%, 45%, 32%, 24% and 50% respectively (Fig. 5 f-l). The 398 improvement for South China (R5) is the highest which is in consistent with the 399 results of active aerosol effects on clouds there. Even in China North Plain (R4) 400 where only two precipitation events occur and last for only 4 days, the mean 401 threat score of T3 is still higher than that of T1 and T2 (Fig.5 c, i). The monthly 402 mean threat scores for T2 are not higher but slightly lower than that for T1 in R2, 403 R4 and R5, indicating that without the real aerosol information from CUACE, the 404 two-moment scheme cannot improve the model precipitation simulation. There is 405 an exception in R3 where threat score of T2 is almost the same with that of T3. 406 This is because that the precipitation is mainly formed by icy clouds in North China 407 (R3) in January and little aerosols can be activated into cloud droplets to 408 participate in the cloud processes, resulting a small threat scoring difference 409 between T2 and T3. It can also indicate that the WDM6 does improve the 410 microphysics for cold cloud formation as the threat scores by T2 and T3 are both 411 higher than that by T1 in R3. 412

To evaluate the overall performance, false alarm or missing events need to be considered. The monthly biases of precipitation simulation, namely (hits + false alarm)/(hits + misses), which infers the over- (larger than 1) or under- (less than 1)

estimates of the rain frequency, are 0.73, 0.75 and 1.13 for T1, T2 and T3 in R1 416 respectively (Table 2). This means that the underestimation by T1 and T2 has been 417 corrected by adding real aerosol activation in T3. This is also true for cases in R2, 418 R4 and R5. Figure 6 is a very typical precipitation distribution pattern for T1, T2 419 and T3 and shows that the precipitations by T3 are very close to the surface 420 observation in terms of timing and coverage from 08:00 (LST) to 14:00 in Jan. 3 by 421 eye-ball comparison. The threat scores are 0.71 and 0.62 and the biases are 1.35 422 and 1.19 for T3, showing a relatively stable and good simulation. As for T1 and T2, 423 threat scores sharply decrease six hours after 08:00. The value of biases indicates 424 that the threat score decreasing comes from severe underestimation as they are 425 much lower than 1. In Northeast China (R3), biases for T2 and T3 are close to 1 426 and close to each other. This is also consistent with the results of cloud water and 427 the threat score in this region that WDM6 performs well for cold clouds and 428 precipitation than single moment scheme WSM6. 429

430 4.2.2 Threat Scoring for Case Evaluation

Two national precipitation events (case 7 and 8) hit most part of east China in R2 including R3, R4 and R5 and six ones (case 1 to case 6) hit south China in R5 in January 2013 (Table 1).The mean threat score for all the cases is 0.446, 0.246 and 0.287 for T3, T2 and T1 respectively. The mean improvement is about 68.0% and the most extraordinary improvement is 192.6% for case 6. Threat scores for T2 are generally lower than those for T1 by -14.5%. These results are also consistent with the conclusions of regional precipitation scoring evaluation.

Both the time series threat score and the case threat score show that the 438 WDM6 scheme alone cannot improve but decrease the ability of precipitation 439 simulation without the real aerosol activation information in the cloud 440 microphysics even though it is more physically based than the one-moment 441 scheme. Additional errors may have been introduced into the model with the 442 prescribed aerosol number concentrations. Only the WDM6 with the aerosol size 443 and number concentration information from CUACE driven by emission and the 444 microphysics, as in T3, can significantly improve the model precipitation 445

446 simulation ability.

# 447 4.3 Aerosol Effects on Temperature

Results of regional-mean temperature profiles in the five targeting areas above 448 700hPa are almost the same with the NCEP reanalysis (Fig.2 b1-b5). The 449 differences between simulation and NCEP reanalysis are mostly under 700hPa in 450 all of the five regions and decrease with height. Temperature is 10 °C and 5 °C 451 higher for all the three tests (T1-T3) than that of NCEP near the surface in R3 and 452 R4, indicating some problems for GRAPES in North China Plain. In South China (R5) 453 the regional-mean temperature profiles are also almost the same as the NCEP 454 reanalysis, showing a good performance of GRAPES over there. Regional mean 455 difference of temperature for different tests in each region is not very obvious 456 with the highest value of about 0.2 °C near the surface located in south China 457 between T3 and T1. Above the surface or in other regions, the mean temperature 458 difference is not significant (< 0.05 °C). This means that the regional mean 459

temperature changed by the aerosol is not obvious. This is also the case for thegeopotential heights.

The time series of differences in the regional mean temperature biases 462 between T3 & T1 and T2 & T1 in the three typical polluted areas (R3, R4 and R5) in 463 east China at the three layers are shown in Figure 7. T3 can clearly decrease the 464 temperature biases below 700 hPa at most time in all the three regions. The 465 difference magnitude decreases from 1000 hPa to 700 hPa, showing the clear 466 impact of aerosol below 700 hPa in these layers. The largest decrease is about 3 °C 467 near the surface and about 1 °C even at 700hPa in R5 during the precipitation 468 event from January 19-22. The difference is less than 0.5°C in R3 and R4 at most 469 time. It also shows that T2 increases the biases with the largest increase in R5. 470 Even in some time T2 might decrease the biases but with a less magnitude 471 compared that of T3. 472

The above analysis also shows that WDM6 with the activation scheme K2000 can introduce errors to the atmospheric temperature fields because of the missing details of aerosol size and component information. The errors would increase with the intensity and frequency of the precipitations. Same as the conclusion from the threat scoring analysis, only the two moment scheme with the real time aerosol activation can reduce the temperature bias and increase the model ability of precipitation simulation.

480 4.4 Feedbacks on surface aerosol simulation

481 To realistically simulate the aerosol impacts on clouds, a good performance of

482	PM <sub>2.5</sub> simulation, including aerosol sizes and vertical distributions, is critical. Due
483	to limited available observations during the simulation period, only daily average
484	of $PM_{2.5}$ concentrations from GRAPES/CUACE have been interpolated and
485	compared with the observations in thirty two stations from CAWNET which have
486	been sorted into the five targeting regions from R1 to R5. Scatter plots for $PM_{2.5}$
487	simulation for R1, R4 and R5, the nation-wide region and the two regions more
488	significantly affected by aerosols, in Fig. 8a-c show that the daily average particle
489	mass concentrations are in a factor of 2 between the observations and model
490	outputs. This is compatible to the simulation level of particle matters in most
491	models.
	Total correlation coefficients for T1, T2 and T2 in D1 to D5 have been platted in

Total correlation coefficients for T1, T2 and T3 in R1 to R5 have been plotted in 492 Fig. 8d. Correlation coefficients for R1, R2 and R5 are all over 0.54 for all the three 493 tests. Correlation coefficients for R4 are about 0.48 for the three tests. This shows 494 a relative stable and reasonably performance for PM<sub>2.5</sub> over these four regions. 495 Correlations coefficients for R3 are only about 0.2 for the three tests showing a 496 relative poor simulation there. These results are a little higher than the result by 497 Zhou 2012 for the same region(Zhou et al., 2012). Fig. 8d also indicates that the 498 differences of correlation coefficients of the three tests in each region are very 499 small. ACI can increase the correlation coefficient by about 2% in terms of monthly 500 501 mean.

502 5. Conclusions

A comprehensive aerosol-cloud-precipitation interaction model has been

developed under the CMA chemical weather modeling system GRAPES/CUACE.
Simulations with this comprehensive system show that interacting activated
aerosols from CUACE with WDM6 in GRAPES/CUACE clearly increase the total
cloud water, liquid cloud water, number concentration of droplets; decrease the
mean diameter of the droplets. It is found that the ultimate efficiency of aerosol
to clouds and precipitation is controlled by multiple parameters, largely by
self-collection growth rates in high aerosol-loading regions.

Studies show that interacting aerosols can obviously increase the model 511 precipitation performance with a threat scoring improvement from 24% to 48% 512 and correct the obvious underestimation by the control test. It is found the 513 physically based, two-moment cloud physics WDM6 in GRPAES/CUACE can 514 produce rational precipitation results with only realistic interactive aerosol inputs 515 in warm and mixed clouds especially in highly polluted regions. WDM6 can also 516 improve the performance of precipitation in cold clouds compared to the control 517 test with the one-moment scheme WSM6. 518

It is further found out that the aerosol-cloud-precipitation interactions in GRAPES/CUACE also reduce temperature bias as well, especially under 700 hPa, which is in harmony with the fact that most aerosols locate below this layer. Aerosol-cloud interaction can decrease the temperature bias by 3 °C at some times during the precipitation event. The monthly mean impact by aerosol-cloud interaction is only about 0.3 °C. Aerosol-cloud interaction's feedback to the surface aerosol concentration is not significant and can increase the correlation

526 coefficient by about 2% in terms of monthly mean.

527	In this paper, an ACI scheme has been explored mainly in winter time when
528	convection is not strong and large part of precipitation is from stratified clouds
529	that can be resolved by WDM6. As the comprehensive and complex relations from
530	emission to cloud and precipitation in this paper provides a platform to study the
531	aerosol impacts on meso-scale weather system in a much wider spatial-temporal
532	scale, the ACI at different time with different weather pattern should be
533	investigated in the future.

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