

Answers to Referee #1's comments

Major:

The authors attempt to empirically link rainfall and other cloud properties to AOD with the implied underlying mechanism that AOD_{CCN} at cloud base. The use of AOD as a proxy for CCN is not robust and has been discussed at length in the literature for many years (see Shinozuka et al. (2015) and references therein). This particular calculation is mainly problematic because AOD is increased by humidity (Twohy et al., 2009), which is also important to rainfall and the various cloud properties the authors are attempting to link to AOD. The authors show that RH covaries with AOD (Fig. 4a), but this is cast in the light of sources of moisture and pollution covarying, which may be partially true, but ignores the leading order problem with their analysis. The authors are guaranteed a correlation between AOD and CF, rain rate, and so on, just by aerosol swelling and increasing scattering. It has been shown that when these effects are taken into account the correlation between AOD and cloud properties becomes much weaker (Christensen et al., 2017). For the paper to be acceptable to be published the authors must use a different proxy for CCN that doesn't build in the result. One possibility would be for the authors to use something like cloud droplet number concentration (CDNC) instead of AOD. This is also a problematic technique as CDNC may potentially be affected by cloud structure/heterogeneity, which will be different between precipitating and non-precipitating clouds. The authors do use CER in their analysis, but this implicitly builds in variations in liquid water content. For example, in a cloud with a fixed CCN and thus a fixed CDNC the LWC can decrease and decrease the CER. The authors do use MACC reanalysis aerosol data as well. They could instead try and rely on the aerosol mass from this product, which has been shown to have skill in predicting variations in CDNC in previous studies (McCoy et al., 2018; Boucher and Lohmann, 1995; Lowenthal et al., 2004). However, in doing this they need to deal with whether variations in precipitation are driving aerosol.

Overall- the built-in correlation between RH and AOD via swelling and the inability to show causality makes this paper unpublishable in ACP.

The authors treat MERRA2 cloud properties like they are observations. They are not. MERRA2 does NOT ingest cloud properties (McCarty et al., 2016). MERRA2 creates clouds just like any other GCM (Molod et al., 2015) and the cloud fraction is not reflective of the observations (see Fig. 9 of Molod et al. (2015)). For example- on L223 P7 the authors refer to the 3D cloud properties. I believe this is just the MERRA2 cloud properties based on the discussion on L147 P5.

The authors may want to consider consulting a copy editor as some of the statements are hard to parse as written. Because the paper must remove the AOD as a proxy of CCN and remove MERRA2 cloud properties I have not bothered to note all the places that the English needs to be clarified.

The abstract is on the short side. This makes it hard to tell what they are actually doing in the paper. The description is very vague. Things like aerosol and humidity are discussed, but it is unclear precisely what they mean. Is it AOD, aerosol number in the PBL? Is it RH or specific humidity? Is it in the PBL or the free troposphere.

R: To respond fully the comments, the manuscript is completely re-written. The revised manuscript is attached and major changes are briefly summarized here:

1. We agree that the use of AOD has inherent issues as the reviewer pointed out. So in addition to AOD, two indicators, the retrieved CDNC and ultraviolet AI, are also selected in the analysis. CDNC calculated by COT and CER from MODIS, is used to separate the different CCN conditions and verify the results based on AOD; see Section 2.1.3. AI from OMI is used to identify rainfall days having absorbing aerosols versus scattering aerosols; see Section 2.1.2. The uncertainties associated with these indicators are given in Section 6.3.

2. For cloud properties, we use CDNC for CCN and the absolute (instead of relative) humidity for moisture. In addition, the analysis is conducted by dividing the heavy rainfall (larger than 8mm/hour) days into four groups to compare the effects of CCN and moisture individually on the cloud properties; see Section 5.2.

3. The results using MERRA2 reanalysis data are deleted.

4. A section of data interpolation is added; see Section 2.2.1.

Specific comments

1.L65 P2: Complicacy is a word, but I am not sure it's fair to say that the complexity of the clouds alone leads to complexity in the indirect effects. Aerosol processes are also important.

R: The statement is revised; see Lines 70-72, Page 3.

2. L70 P3: The author refers to the Albrecht/lifetime/adjustments and the Twomey effect/ first indirect effect as the Twomey effect.

R: The statement is corrected; see Lines 73-75, Page 3.

3. L74 P3: What is the different condition of moisture? I think what the authors mean is that if the air near cloud is moist or not the cloud droplet size can increase or decrease. This sentence needs a bit of clarification. The Twomey effect is not related to cloud droplet radius. It is the relation between CCN and N_d .

R: The different condition of moisture was referred to different moisture supply near the cloud base. In any case, we have re-written the sentence for clarity; see Lines 80-85, Page 3.

4. L82 P3: In one day of what? Do the authors mean that convective rainfall usually starts and stops within a day?

R: "In one day" refers to that the heavy convective rainfall over BTH region usually occurs within 24 hours, which is no more than 20 hours as shown in Fig.2 in the manuscript. The average duration of all heavy rainfall events is 6.1 hours. We modified the sentence for clarity; see Lines 88-90, Page 3.

5. L111 P4: I assume this means above sea level. Also, wouldn't the orographic effect be the lifting by the orography, so the slope and the advection, not just the height? Limiting to sub-100m is probably ok, but there needs to be a better discussion of this.

R: Thanks for your suggestion. The "below the topography of 100 meter" does mean above sea level. We just want to focus on the aerosol impact on the rainfall diurnal variation in the plain area in order to reduce the probable influence of orography lifting effect like you said on the rainfall diurnal variation as far as possible. Below 100 meters above the sea level is a simple criterion for choosing plain area, and in the study we temporarily don't consider other orography effects. We added a figure of orography and stations over the study region to show that we select the plain area; see Lines 123-126, Page 4.

6. L114 P4: AOD is not a proxy for particle number. It is the brightness of aerosol in the column of atmosphere, which in turn is a function of the scattering of the particles, the number of particles, and the mass of particles. The authors want to relate this quantity to CCN, which is a very outdated idea. CCN is more directly related to AI, but even then this is a highly imperfect metric because it is sensitive to aerosol swelling by moisture and the vertical structure. See, for example, Shinozuka et al. (2015). The use of AOD as a proxy for CCN in this paper is a critical error and makes it hard to draw any meaningful conclusions from their analysis because AOD is enhanced by RH (Christensen et al., 2017; Twohy et al., 2009).

R: Yes, we agree, CDNC is also used as another indicator.

7. L134 P4: The spatial resolution is not $1^{\circ} \times 1^{\circ}$ in MACC. The authors presumably mean the data is regridded to $1^{\circ} \times 1^{\circ}$.

R: The MACC reanalysis data we downloaded is the version of retrieve NetCDF with spatial resolution of $1^{\circ} \times 1^{\circ}$ which is the same as the resolution of MODIS (the resolution of ECMWF data is optional, see <http://apps.ecmwf.int/datasets/data/macc-reanalysis/levtype=sfc>). To unify the datasets, we interpolated the gridded datasets onto stations using the average value in a $1^{\circ} \times 1^{\circ}$ grid as the background condition of each rainfall station; see Section 2.2.1.

8. L141 P4: Provide a citation for the evaluation of MODIS CTH with CALIPSO.

R: References are added; see Lines 159, Page 5.

9. L147 P5: The authors appear to be using cloud properties from MERRA2 as if they were observations. This is a model product and is not nudged to agree with observations in any way. This is another critical flaw in the paper.

R: We agree. The results using MERRA2 data are deleted.

10. L159 P5: The ECMWF-Interim resolution is not $1 \times 1^\circ$ - again I assume this is the gridded resolution used in the study, but is confused by saying MERRA2 is $0.624 \times 0.5^\circ$ (L152).

R: Same with MACC reanalysis data, ERA-interim data we downloaded is also the version of retrieve NetCDF with spatial resolution of $1^\circ \times 1^\circ$. The horizontal resolution of MERRA2 data we downloaded is $0.624^\circ \times 0.5^\circ$ (now we deleted the part of MERRA2), and then we interpolated all the gridded datasets onto rainfall stations using the average value in a $1^\circ \times 1^\circ$ grid as the background condition of each rainfall station; see Section 2.2.1.

11. L184 P6: How was the t-test applied? Was it difference in means or difference in the means within a bin. Throughout the paper it is stated that results are significant at 95% confidence, but it is unclear what this actually means.

R: Student's t-test is used to check the significance of differences in means between different groups not within a bin. And the numbers of bins in the study have been all tested for better representing the PDF distribution. We have re-written the sentence for clarity; see Lines 235-236, Page 7.

12. L223 P7: MERRA2 is NOT equivalent to observations of cloud properties. This is just an analysis of the modeled cloud properties in MERRA2.

R: We agree, this part is deleted.

13. L239 P8: Different moisture conditions (which I guess just means RH at a fixed pressure level) also affect clouds.

R: Yes, they are more explicit now; see Lines 331-332, Page 10.

14. L240 P8: Fixing the wind direction is not enough to orthogonalize RH, AOD, and clouds.

R: We perceive that the circulation, radiation, moisture condition and cloud all influence rainfall. However, changes of radiation and cloud may associate more directly with aerosols. We fix the wind direction to remove circulation influence. To investigate the influence of moisture supply on aerosol cloud effect, we have additionally divide the heavy rainfall days into four groups and compare the influences of CCN and moisture individually on the cloud properties; see Section 5.2. Nevertheless, the sentence is modified; see Lines 332-333 of Page 10.

15. L300 P9: The authors do try and use BC and SO₄ mass concentrations, which sort of gets around the issues with AOD and RH. The model level that the concentration is taken at is not specified. However, the authors still partition by AOD, and just use BC and SO₄ to partition the high and low AOD cases. The results shown in Fig. 8 are not terribly convincing. It is stated that the results are significant at 95% confidence. What does this mean? Are the means different and 95% confidence or are the values of the

PDF in each bin different at 95% confidence? From just looking at the PDFs it seems like only the BC peak time and SO₄ duration are all that are different. The CF PDFs in Fig. 9 look different for SO₄, but the jump in the PDF of CF at 90% does not look terribly robust. This may change if more bins are used in the PDF.

R: To respond to the comments, the ultraviolet AI from OMI is also used, which further confirm the findings. The significance refers to the significance of difference in means rather than in each bin. As shown in Fig. A1 below, the results are consistent for different bins. A statement is therefore added; see Lines 234-235 of Page 7.

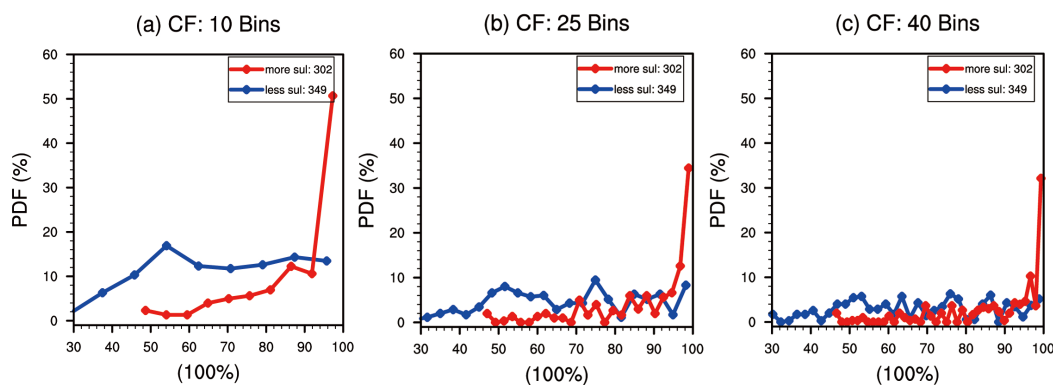


Figure A1. PDF of CF (units: %) in different conditions of sulfate using (a) 10 bins, (b) 25 bins and (c) 40 bins. Blue/red lines stand for the condition of less/more sulfate.

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1 **An observational study of the effects of aerosols on diurnal variation of heavy rainfall**
2 **and the concurrent cloud changes over Beijing-Tianjin-Hebei**

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36 **Abstract:** Our recent observational study found that the rainfall diurnal variation over Beijing-Tianjin-Hebei
37 shows distinct signature of the effects of pollutants. Here we used the hourly rainfall measurements together
38 with daily satellite-based information of aerosols and clouds to further study the responses of heavy rainfall
39 and cloud properties to increases of pollutants. While the aerosol optical depth (AOD) and cloud droplet
40 number concentration (CDNC) are both used as pollution indicators to provide a different perspective in
41 rainfall study, CDNC is used exclusively on cloud changes. It is found that both indicators yield three
42 consistent and distinguished responses of heavy rainfall: earlier start time, earlier peak time, and longer
43 duration. However, quantitative differences exist between the two: for the first two responses, the advances
44 are 0.7 and 1.0 hours respectively with AOD, but 2.1 and 4.2 hours respectively with CDNC; the third is
45 prolonged by 0.8 hours with AOD and 2.4 hours with CDNC. In-depth analysis suggests that earlier in both
46 start time and peak time occur in the presence of absorbing aerosols while the longer duration is attributed to
47 scattering aerosols. Changes in cloud statistics caused by aerosols show increases in cloud fraction (11.1%),
48 cloud top pressure (37.8 hPa), the liquid/ice cloud optical thickness (32.2/26.0) and cloud water path
49 (239.8/422.0 g/m²); and decreases in liquid/ice cloud effective radius (8.6/8.7μm). Analyses also indicate that
50 increased moisture tends to decrease the cloud top pressure and enlarge the liquid cloud effective radius,
51 which partially compensate the aerosol effects. Finally, the mechanisms accounted for the aerosol effects on
52 heavy rainfall are hypothesized.

53 **Key words: aerosol, heavy rainfall, diurnal variation, cloud, Beijing-Tianjin-Hebei, observational study**

54

55 **1. Introduction**

56 Aerosols modify the global hydrologic cycle through both radiative effect (direct effect) and cloud effect
57 (indirect effect) (IPCC, 2013). On the one hand, through absorbing or scattering solar radiation, aerosols can
58 lead to the air aloft heating (e.g. Jacobson 2001; Lau et al. 2006) or the surface cooling (Lelieveld and
59 Heintzenberg 1992; Guo et al. 2013; Yang et al., 2018), which changes the atmospheric vertical static stability
60 and modulates rainfall (e.g. Rosenfeld et al. 2008). On the other hand, water-soluble aerosols serving as cloud
61 condensation nuclei (CCN) could affect the warm-rain processes and cold-rain processes through influencing
62 the cloud droplet size distributions, cloud top heights and other cloud properties (Jiang et al., 2002; Givati and
63 Rosenfeld 2004; Chen et al., 2011; Lim and Hong 2012; Tao et al., 2012). Beijing-Tianjin-Hebei (BTH) region
64 is the heaviest aerosol polluted area in China and concerns have been raised about the
65 aerosol-radiation-cloud-precipitation interaction over this region. The impact of aerosols on light rainfall or
66 warm-rain processes over BTH region almost reaches consistent agreement (e.g., Qian et al., 2009), but
67 aerosol effects on the heavy convective rainfall in this region still have large uncertainties (Wang et al., 2009;
68 Guo et al., 2014; Wang et al., 2016).

69 The clouds that can generate heavy convective rainfall in BTH region usually contain warm clouds, cold

70 clouds and mixed-phase clouds (e.g. Guo et al., 2015). Because the aerosol-cloud interactions in different
71 types of clouds are distinct (Gryspeerd et al., 2014), aerosol indirect effect during heavy rainfall is more
72 complicated than its direct effect (Sassen et al., 1995; Sherwood, 2002; Jiang et al., 2008, Tao et al., 2012).
73 For warm clouds, by serving as CCN for more cloud droplets, aerosols can increase cloud albedo so called albedo
74 effect or Twomey effect (Twomey, 1977), lengthen the cloud lifetime so called lifetime effect (Albrecht, 1989), and
75 enhance thin cloud thermal emissivity so called thermal emissivity effect (Garrett and Zhao, 2006). The above
76 effects tend to increase the cloud microphysical stability and suppress warm-rain processes (Albrecht 1989;
77 Rosenfeld et al. 2014). For cold clouds and mixed-phase clouds, many studies reported that the cloud liquid
78 accumulated by aerosols is converted to ice hydrometeors above the freezing level, which invigorates deep
79 convective clouds and intensifies heavy precipitation so called invigoration effect (Rosenfeld and Woodley,
80 2000; Rosenfeld et al., 2008; Lee et al. 2009; Guo et al. 2014). The Twomey effect infers that aerosols serving
81 as CCN increasing the cloud droplets could reduce cloud droplet size within a constant liquid water path
82 (Twomey, 1977). However, the opposite results of relationship between aerosols and cloud droplet effective
83 radius were reported in observations (Yuan et al., 2008; Panicker et al., 2010; Jung et al., 2013; Harikishan et
84 al., 2016; Qiu et al., 2017), which might be related with the moisture supply near the cloud base (Yuan et al.,
85 2008; Qiu et al., 2017). Besides, the influence of aerosols on ice clouds also depends upon the amount of
86 moisture supply (Jiang et al., 2008). Therefore, how the aerosols modify the clouds associated with heavy
87 convective rainfall does not reach a consensus, particularly if considering the different moisture conditions.

88 Heavy convective rainfall over BTH region usually occurs within a few hours, thus studying on the
89 relationship between aerosols and rainfall diurnal variation could deepen our understanding of aerosol effects
90 on heavy rainfall. Several previous studies have found that aerosols are related to the changes of the rainfall
91 diurnal variation in other regions (Kim et al., 2010; Gryspeerd et al., 2014; Fan et al., 2015; Guo et al., 2016;
92 Lee et al., 2016). However, the above studies do not address the change of cloud properties and its sensitivity
93 to different conditions of moisture supply. Although our recent work over BTH region (Zhou et al. 2018)
94 attempted to remove the meteorological effect including circulation and moisture and found that the peak of
95 heavy rainfall shifts earlier on the polluted condition, it only excluded the extreme moisture conditions and
96 focused on aerosol radiative effect on the rainfall diurnal variation. Therefore, this study aims to deepen the
97 previous study (Zhou et al., 2018) through investigating the following questions: (1) how do aerosols modify
98 the different features of the diurnal variation of heavy rainfall (start time, peak time, duration and intensity)?
99 (2) how do different types of aerosols (absorbing aerosols and scattering aerosols) modify the characteristics
100 of heavy rainfall? (3) how do aerosols influence the concurrent cloud properties with inclusion of moisture?
101 To solve above questions, we used aerosol optical depth (AOD) as an indicator of pollution to compare the
102 characteristics of heavy rainfall, used cloud droplet number concentration (CDNC) representing CCN to
103 investigate the changes of rainfall and clouds, and used aerosol index (AI) to distinguish the different effects
104 of absorbing aerosols and scattering aerosols. The paper is organized as following: The data and methodology
105 are introduced in Sect. 2. Section 3 presents the distinct characteristics of rainfall diurnal variation on

106 clean/polluted conditions using AOD and CDNC. Section 4 addresses the impacts of different types of
107 aerosols on characteristics of heavy rainfall. Section 5 describes the changes of cloud properties with the
108 increase of CCN and moisture. Section 6 discusses the aerosol effects on cloud with inclusion of moisture, the
109 distinct roles of aerosol radiative effect and cloud effect in heavy rainfall and the uncertainties of different
110 indicators. Conclusion will be given in Sect. 7.

111

112 **2. Data and methodology**

113 **2.1 Data**

114 Four types of datasets from the year 2002 to 2012 (11 years) were used in this study, which include (1)
115 precipitation, (2) aerosols, (3) clouds, and (4) other meteorological fields.

116 **2.1.1 Precipitation data**

117 To study the diurnal variation of heavy rainfall, the gauge-based hourly precipitation datasets were used,
118 which were obtained from the National Meteorological Information Center (NMIC) of the China
119 Meteorological Administration (CMA) (Yu et al., 2007) at 2420 stations in China from 1951 to 2012. The
120 quality control made by CMA/NMIC includes the check for extreme values (the value exceeding the monthly
121 maximum in daily precipitation was rejected), the internal consistency check (wiping off the erroneous
122 records caused by incorrect units, reading, or coding) and spatial consistency check (comparing the time series
123 of hourly precipitation with nearby stations) [Shen et al., 2010]. Here we chose 176 stations in the plain area
124 of BTH region that are below the topography of 100 meter above sea level as shown in Fig.1, which is
125 consistent with our previous work because we purposely removed the probable orographic influence on the
126 rainfall diurnal variation (Zhou et al., 2018). The record analyzed here is the period of 2002 to 2012.

127 **2.1.2 Aerosol data**

128 AOD is a proxy for the optical amount of aerosol particles in a column of the atmosphere and serves as one of
129 indicators for the division of aerosol pollution condition in this study, was obtained from MODIS (Moderate
130 Resolution Imaging Spectroradiometer) Collection 6 L3 aerosol product with the horizontal resolution of
131 $1^\circ \times 1^\circ$ onboard the Terra satellite (Tao et al., 2015). The quality assurance of marginal or higher confidence
132 was used in this study. The reported uncertainty in MODIS AOD data is on the order of (-0.02-10%),
133 (+0.04+10%) (Levy et al., 2013). The Terra satellite overpass time at the equator is around 10:30 local solar
134 time in the daytime, which is before the occurrence of heavy rainfall events in this study as shown in Fig. 2.
135 Therefore, the AOD used here represents the situation of the air quality in advance of heavy rainfall
136 appearance.

137 The ultraviolet AI from Ozone Monitoring Instrument (OMI) on board the Aura satellite which was
138 launched in July 2004 is used for detecting the different types of aerosols in this study. The OMI ultraviolet

139 AI is a method of detecting absorbing aerosols from satellite measurements in the near-ultraviolet wavelength
140 region (Torres et al., 1998). The positive values of ultraviolet AI are attributed to the absorbing aerosols such
141 as smoke and dust while the negative values of AI stand for the non-absorbing aerosols (scattering aerosols)
142 such as sulfate and sea salt (Tariq and Ali, 2015). The near-zero values of AI occur when clouds and Rayleigh
143 scattering dominate (Hammer et al., 2018). The horizontal resolution of AI data is $1^\circ \times 1^\circ$ and it covers the
144 period of 2005 to 2012.

145 MACC-II (Monitoring Atmospheric Composition and Climate Interim Implementation) reanalysis product
146 produced by ECMWF (the European Centre for Medium-Range Weather Forecasts), provided the AOD
147 datasets for different kinds of aerosols (BC, sulfate, organic matter, mineral dust and sea salt). MACC-II
148 reanalysis products are observationally-based within a model framework, which can offer a more complete
149 temporal and spatial coverage than observation and reduce the shortcomings of simulation that fail in
150 simulating the complexity of real aerosol distributions (Benedetti *et al.*, 2009). The horizontal resolution of
151 MACC-II is also $1^\circ \times 1^\circ$ with the time interval of six-hour. MACC-II data covers the period of 2003 to 2012.

152 2.1.3 Cloud data

153 Daily cloud variables, including cloud fraction (CF), cloud top pressure (CTP), cloud optical thickness (COT,
154 liquid and ice), cloud water path (CWP, liquid and ice) and cloud effective radius (CER, liquid and ice), were
155 obtained from MODIS Collection 6 L3 cloud product onboard the Terra satellite. The MODIS cloud product
156 combines infrared emission and solar reflectance techniques to determine both physical and radiative cloud
157 properties (Platnick et al., 2017). The validation of cloud top properties in this product has been conducted
158 through comparisons with CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) data and other lidar
159 observations (Holz et al., 2008; Menzel et al., 2008), and the validation and quality control of cloud optical
160 products is performed primarily using in situ measurements obtained during field campaigns as well as the
161 MODIS Airborne Simulator instrument (<https://modis-atmos.gsfc.nasa.gov/products/cloud>). Since the clouds
162 associated with heavy rainfall in the BTH region during early summer contain warm clouds, cold clouds and
163 mixed-phase clouds (e.g. Guo et al., 2015), we purposely selected the clouds with its top pressure above 600
164 hPa to investigate both liquid and ice cloud properties because the 0°C isotherm of BTH region is nearly
165 located at this height. Consistent with AOD, the measure of above cloud variables is before the occurrence of
166 heavy rainfall.

167 CDNC is retrieved as the proxy for CCN and also another indicator for separating different aerosol
168 conditions in this study. Currently, most derivations of CDNC assume that the clouds are adiabatic and
169 horizontally homogeneous; CDNC is constant throughout the cloud's vertical extent, and cloud liquid water
170 content varies linearly with altitude adiabatically (Min et al., 2012; Bennartz and Rausch, 2017). According to
171 Boers et al. (2006) and Bennartz (2007), we calculated CDNC (unit: cm^{-3}) through:

$$172 \quad \text{CDNC} = \frac{C_W^{1/2}}{k} \frac{10^{1/2}}{4\pi\rho_w^{1/2}} \frac{\tau^{1/2}}{R_e^{5/2}} \quad (1)$$

173 Where C_w is the moist adiabatic condensate coefficient, and its value depends slightly on the temperature of
 174 the cloud layer, ranging from 1 to $2.5 \times 10^{-3} \text{ gm}^{-4}$ for a temperature between 0 °C and 40 °C (Breguier,
 175 1991). In this study, we calculated the C_w through the function of the temperature (see Fig.1 in Zhu et al.,
 176 2018) at a given pressure that is 850 hPa. And we have tested the sensitivity of CDNC to the amount of C_w
 177 and found it almost keeps the same when the C_w changes from 1 to $2.5 \times 10^{-3} \text{ gm}^{-4}$. The coefficient k is the
 178 ratio between the volume mean radius and the effective radius and varies between 0.5 and 1 (Breguier et al.,
 179 2000). Here we used $k = 1$ for that we cannot get the accurate value of k and the value of k does not influence
 180 the rank of CDNC for the division of aerosol condition in this study. ρ_w is cloud water density. τ and Re are
 181 the COT and CER obtained from MODIS Collection 6 L3 cloud product onboard the Terra satellite.

182 2.1.4 Other meteorological data

183 Other meteorological factors, including wind, temperature, pressure and specific humidity, were obtained
 184 from the ERA-Interim reanalysis datasets with $1^\circ \times 1^\circ$ horizontal resolution and 37 vertical levels at six-hour
 185 intervals. ERA-Interim is the global atmospheric reanalysis produced by ECMWF, which covers the period
 186 from 1979 to near-real time (Dee et al., 2011). The absolute humidity (AH), which stands for the water vapor
 187 content of air per unit volume, is calculated as the indicator of moisture supply in this study. We calculated the
 188 AH (unit: g/m^3) through:

$$189 \quad AH = \frac{1000e}{R_v T} \quad (2)$$

190 Where R_v is the specific gas constant, which is $461.5 \text{ J kg}^{-1} \text{ K}^{-1}$. T is air temperature (unit: K), and the vapor
 191 pressure e (unit: hPa) is calculated by the equation below:

$$192 \quad q = \frac{0.622e}{P - (1 - 0.622)e} \quad (3)$$

193 Where q is specific humidity (unit: kg/kg) and P is atmosphere pressure (unit: hPa), which were both obtained
 194 from ERA-interim.

195

196 2.2 Methodology

197 2.2.1 Method of interpolation

198 We used both station data of gauge-based precipitation and gridded data including aerosols, clouds and other
 199 meteorological variables. Gridded datasets in this study were downloaded with the horizontal resolution of
 200 $1^\circ \times 1^\circ$, which are consistent with the resolution of MODIS L3 product. To unify the datasets, we interpolated
 201 all the gridded datasets onto the selected 176 rainfall stations using the average value in a $1^\circ \times 1^\circ$ grid as the
 202 background condition of each rainfall station, i.e., the stations in the same $1^\circ \times 1^\circ$ grid have the same aerosol,
 203 cloud and meteorological conditions.

204 2.2.2 Selection of sub-season and circulation

205 Consistent with our previous work, we focused on early summer (1 June to 20 July) before the large-scale
206 rainy season starts, in order to remove the large-scale circulation influence and identify the effect of aerosols on
207 local convective precipitation because BTH rainfall during this period is mostly convective rainfall (Yu et al.,
208 2007) with heavy pollution (Zhou et al., 2018). And to unify the background atmospheric circulation, we only
209 selected the rainfall days with southwesterly flow, which is the dominant circulation accounting for 40% of
210 total circulation patterns over the BTH region during early summer (Zhou et al., 2018).

211 2.2.3 Classification of the heavy rainfall, clean/polluted and moisture conditions

212 With the circulation of southwesterly, we selected heavy rainfall days when the hourly precipitation amount
213 was more than 8.0 mm/hour (defined by *Atmospheric Sciences Thesaurus, 1994*). We used two indicators to
214 distinguish the clean and polluted condition, which are AOD and CDNC. The 25th and 75th AOD/CDNC of the
215 whole rainfall days are used as the thresholds of clean and pollution condition, and the values are shown in
216 Tab.1. It shows that there are 514 cases of heavy rainfall on polluted days and 406 cases of that on clean days
217 when using AOD, and 924/894 cases on polluted/clean condition when using CDNC.

218 The absorbing aerosols are detected using the positive values of AI that is named as absorbing aerosol index
219 (AAI) here, and we can retrieve the scattering aerosol index (SAI) using the negative values of AI. AAI and
220 SAI are also divided into two groups using the threshold of 25th/75th as shown in Tab.1. We used AAI/SAI
221 more than 75th as the extreme circumstances of absorbing aerosols and scattering aerosols to compare their
222 impacts on heavy rainfall. The case numbers are 375 and 550 events respectively for the extreme AAI and
223 SAI cases. Using the same method, we chose cases of more BC/sulfate when the AOD of BC/sulfate is larger
224 than the 75th AOD of itself in all rainy days, and cases of less BC/sulfate when that is less than the 25th AOD
225 of itself in the same condition. Accordingly, we selected 459 cases of more BC and 274 cases of less BC with
226 heavy rainfall. Similarly, 361 cases of more sulfate and 419 cases of less sulfate with heavy rainfall were
227 selected.

228 The AH at 850 hPa is used as the indicator of moisture supply. We chose wet cases when the AH on that
229 rainy day is larger than 75th percentile of the whole rainy days, and chose dry cases when AH on that day is
230 less than the 25th percentile of the whole rainy days (the thresholds are shown in Tab. 1).

231 2.2.4 Statistical analysis

232 We adopted the probability distribution function (PDF) to compare the features of heavy rainfall and cloud
233 variables on different conditions of aerosols, through which we can understand the changes of rainfall/cloud
234 properties more comprehensively. The numbers of bins we selected in the study have been all tested for better
235 representing the PDF distribution. Student's t-test is used to examine the significance level of differences
236 between the different groups of aerosol conditions.

237

238 **3 Distinct characteristics of heavy rainfall diurnal variation associated with aerosol pollution**

239 Our previous study (Zhou et al. 2018) has reported the distinct peak shifts of rainfall diurnal variation between
240 clean and polluted days using the indicator of AOD over the BTH region during early summer. Similar with
241 our previous study, the PDF of the heavy rainfall peak time shows that the maximum of rainfall peak is about
242 two hours earlier on the polluted days (20:00 LST) than that on the clean days (22:00 LST) (Fig. 2a(2)). To
243 comprehensively recognize the changes of rainfall diurnal variation associated with air qualities, here we
244 examined the PDF of the start time, the duration and the intensity besides the peak time of heavy rainfall.

245 **In terms of the start time of heavy rainfall, a significant advance is found as shown in Fig. 2a(1). The**
246 **secondary peak on the early morning is ignored here because the early-morning rainfall might be associated with**
247 **the mountain winds (Wolyn et al., 1994; Li et al., 2016) and the nighttime low-level jet (Higgins et al., 1997; Liu et**
248 **al., 2012) that is beyond the scope of this study.** The time for maximum frequency of heavy rainfall initiation
249 is 6 hours earlier on the polluted days, shifting from around 0:00 LST on the clean days to the 18:00 LST.
250 Regarding the rainfall durations, the average persistence of heavy rainfall on polluted days is 0.8 hours longer
251 than that on clean days (Tab. 2). According to the PDF shown as in Fig. 2a (3), the occurrence of short-term
252 precipitation (≤ 6 hours, Yuan et al., 2010) decreases while that of long-term precipitation (> 6 hours, Yuan et
253 al., 2010) increases. The intensity of hourly rainfall exhibits a decrease on the polluted days. However,
254 compared with the other features, the change of intensity does not pass the 95% statistical confidence level.

255 **The differences of rainfall characteristics between clean and polluted days above can be well detected using**
256 **the indicator of AOD. Since this study would investigate the aerosol-cloud interaction, the property of aerosol**
257 **acting as CCN should be emphasized. In this condition, we did the similar analysis to verify the results above**
258 **using the retrieved CDNC as the indicator of CCN (Zeng et al., 2014; Zhu et al., 2018) since AOD is not a**
259 **proper proxy for CCN (Shinozuka et al., 2015). As a result, the same phenomenon can be well exhibited as**
260 **shown in Fig. 2b. The start time and peak time of the heavy rainfall on polluted condition also show**
261 **significant advances compared with the clean condition, with the average advances of 2.1 hours and 4.2 hours**
262 **respectively (Tab. 2). The duration of heavy rainfall on the polluted condition is also prolonged, which is 2.4**
263 **hours longer in average (Tab. 2). Similar with the results based on AOD, the difference of rainfall intensity**
264 **between clean and polluted conditions using CDNC does not pass the 95% statistical confidence level as well.**

265 **Both results of AOD and CDNC show that the start and peak time of heavy rainfall occur earlier and the**
266 **duration becomes longer under pollution, although the quantitative differences exist between the two**
267 **indicators. Since the difference of rainfall intensity is not significant, the following study only focuses on**
268 **investigating why the start time, peak time and duration of heavy rainfall change with pollution in the diurnal**
269 **time scale.**

270

271 **4 Impacts of different aerosols on rainfall diurnal variation**

272 Using the indicator of AI, we further investigate the different changes of rainfall characteristics related to
273 absorbing aerosols and scattering aerosols respectively. The PDF of start time, peak time and duration of
274 heavy rainfall under the extreme circumstances of absorbing aerosols and scattering aerosols are compared in
275 Fig. 3. The rainfall start time on absorbing aerosol days shows a significant advance with the maximum
276 frequency occurring at 20:00 LST, compared with the 3:00 LST on scattering aerosol days (Fig. 3a). Similarly,
277 the rainfall peak time also shows earlier on absorbing aerosol days, with an average advance of 1.7 hours (Fig.
278 3b). The rainfall duration on scattering aerosol days shows longer than that on absorbing aerosol days, which
279 are 5.9 hours and 5.0 hours respectively in average. All the differences above between the two groups have
280 passed 95% statistical confidence level. The results indicate that the absorbing aerosols and scattering aerosols
281 may have different or inverse effect on heavy rainfall that absorbing aerosols may generate the heavy rainfall
282 in advance and the scattering aerosols may delay and prolong the heavy rainfall.

283 To further distinguish the effects of the absorbing/scattering aerosols on the heavy rainfall, we purposely
284 re-examine the above findings through BC/sulfate that can represent typical absorbing/scattering aerosols over
285 BTH region. BC has its maximum center over BTH region (Fig. 4a) and our previous study has indicated that
286 the radiative effect of BC low-level warming may facilitate the convective rainfall generation (Zhou et al.,
287 2018). The percentage of sulfate is also large in BTH region (Fig. 4b) and the sulfate is one of the most
288 effective CCN that influences the precipitation in this region (Gunthe et al., 2011). Accordingly, we selected
289 the cases with different amounts of BC and sulfate AOD to compare the role of them in the diurnal variation
290 of heavy rainfall. The methods have been described in Sect. 2.2.3. The PDF of the start time, peak time and
291 duration of heavy rainfall were shown for the higher and lower BC cases in Fig. 5a, respectively. The most
292 striking result is that the maximum frequency of rainfall start time in high BC cases evidently shifts earlier by
293 7 hours from 19:00 LST to 2:00 LST. Meanwhile, compared with low BC cases, the mean peak time in high
294 BC cases shows 1.0 hour earlier than that in low BC cases. And the duration of heavy rainfall is slightly
295 shorter in high BC cases with the mean difference of 0.2 hours. These features of higher BC cases are
296 consistent with the above absorbing aerosol effect. In contrast, when the sulfate has higher amount, the mean
297 start time of rainfall is delayed by 0.5 hours, while the duration shows a significant increase by 1.5 hours in
298 average. The behaviors of higher sulfate cases exhibit similar with the above scattering aerosol effect (Fig.
299 5b).

300

301 **5 Cloud effect of aerosols with inclusion of moisture**

302 **5.1 Characteristics of clouds on clean and polluted condition based on CDNC**

303 To understand the cloud effect of aerosols during heavy rainfall, we need to recognize the concurrent cloud
304 characteristics on clean and polluted conditions. **The cloud properties we used were obtained from satellite**

305 product which were measured at the same time as aerosols before the occurrence of heavy rainfall. The
306 differences of cloud features were examined in both macroscopic properties (including CF, CTP, COT and
307 CWP) and microscopic properties (including CER) between the clean and polluted condition based on CDNC,
308 as shown in Fig. 6. The PDF distribution of CF shows that the CF on the polluted condition is evidently larger
309 than that on the clean condition. The average CF is 82.5% on the clean condition and 93.6% on the polluted
310 condition. The average CTP on the polluted condition is 436.0 hPa, more than that on the clean condition
311 which is 398.2 hPa, indicating that the cloud top height is lower on the polluted days. According to PDF
312 distribution, CTP on polluted condition has a significant peak at around 300 hPa and secondary maximum at
313 around 550 hPa.

314 The COT, CWP and CER were further analyzed for the liquid and ice portions of clouds as shown in Fig. 6.
315 Both liquid and ice COT on polluted condition exhibit a significant increase compared with that on clean
316 condition. The mean amount of liquid COT increases by 32.2 and ice COT increases by 26.0. Similar with
317 COT, the amount of liquid and ice CWP also increase on polluted condition. And the mean amount of liquid
318 CWP increases by 239.8 g/m² and ice CWP increases by 422.9 g/m². The PDF of liquid CER on the polluted
319 condition shows a shift to the smaller size and its mean value decreases by 8.6 μm. In accordance with the
320 CER of liquid clouds, the CER of ice clouds also shows decrease with the mean difference of 8.7 μm. The
321 differences of above cloud properties between clean and polluted cases have all passed the 95% statistical
322 confidence level.

323 According to the above results, the increased CDNC corresponds to the increase of CF, COT, CWP for both
324 liquid and ice clouds, but the decrease of cloud top height and CER (liquid and ice). Since we cannot
325 distinguish the liquid part of mix-phased clouds from liquid (warm) clouds in the observation, the changes of
326 liquid cloud properties above might come from both the liquid (warm) clouds and the liquid part of
327 mixed-phase clouds. Likewise, the above-mentioned changes of ice cloud properties might come from both
328 ice (cold) clouds and the ice part of mixed-phase clouds.

329

330 **5.2 Influence of CCN and moisture on cloud properties**

331 The different moisture supply under the cloud base can influence the cloud properties as well as the effect of
332 aerosols on cloud properties (Yuan et al., 2008; Jiang et al., 2008; Jung et al., 2013; Qiu et al., 2017). It is hard
333 to completely remove the moisture effect on the above results in a pure observational study. Since the
334 southwesterly circulation cannot only transport pollutants but also moisture to the BTH region (Wu et al.,
335 2017), more pollution usually corresponds to more moisture (Sun et al., 2015). Because the moisture supply
336 for BTH is mainly transported via low-level southwesterly circulation, we purposely used the AH at 850 hPa
337 as the indicator of moisture condition. To identify the effect of aerosols on clouds and its sensitivity to
338 moisture, we purposely investigated the changes of above cloud properties with different conditions of CDNC

339 and moisture respectively. We categorized all cases of heavy rainfall into four groups, which are (1) clean and
340 dry, (2) polluted and dry, (3) clean and wet, (4) polluted and wet, and checked the changes of above cloud
341 properties, as shown in Tab. 3. Here “clean/polluted” refers to the CDNC on that rainfall day less/more than
342 25th/75th percentile of the CDNC among the whole rainfall days, and similarly, the “dry/wet” refers to the AH
343 on that rainfall day less/more than 25th/75th percentile of itself among the whole rainfall days. We made the
344 significant test of differences between group 1 and 2, group 1 and 3, group 2 and 4, group 3 and 4.

345 Comparing the results of group 1 and 2, which are both on the dry condition, we can identify the influence
346 of CDNC on cloud properties. The changes of these cloud variables are the same as that in Sect. 5.1, that the
347 CF, COT and CWP both for liquid and ice are increased on the polluted condition, while the cloud top height
348 and liquid and ice CER are decreased. Among these variables, the COT and CWP both for liquid and ice are
349 especially larger on polluted condition, which are 2-5 times larger than that on clean condition. The liquid
350 CER on polluted condition also changes evidently, which becomes almost a half of that on clean condition.
351 On the wet condition, comparing the results of group 3 and 4, the changes are also similar that liquid and ice
352 CER are decreased and others are increased except that the change of CTP is not significant. The results of the
353 two comparisons above indicate that with the increase of CDNC (CCN), the CF, COT, CWP are increased
354 while the CER is decreased regardless of the moisture amount.

355 Comparing the results of group 1 and 3, we can get the changes of cloud properties related only to moisture
356 on the same clean condition. A common feature is that CF, cloud top height, COT and CWP both for liquid
357 and ice clouds exhibit increases along with the increase of AH (the decrease of CTP corresponds to the
358 increase of cloud top height). Compared with the CF on clean and dry condition (group 1), the increase of CF
359 on clean and wet condition (group 3) is larger than that on polluted and dry condition (group 2), which
360 indicates the influence of moisture on CF might be larger than the influence of CCN. In contrast with CF, the
361 increases of COT and CWP both for liquid and ice clouds in group 2 are 2-3 times larger than that in group 3,
362 which indicates that the influences of moisture on COT and CWP are evidently smaller than the influence of
363 CCN. The influences of moisture on liquid and ice CER are not significant on the same clean condition. On
364 polluted condition, comparing group 2 and 4, we found the same changes are the increase of CF, liquid COT
365 and CWP, and the decrease of CTP, while the influences of moisture on ice COT and CWP on the polluted
366 condition become not significant. When the moisture increases, the liquid CER on polluted condition is
367 increased and the ice CER is decreased.

368 The results above indicate that both CCN and moisture have impacts on cloud properties. They both
369 contribute to the increase of CF, COT and CWP, in which the influence of CCN on CF is smaller but its
370 influences on COT and CWP are larger than moisture. The CCN and moisture have opposite effects on CTP,
371 that the moisture can decrease the CTP which is lifting the cloud top, while CCN can lower the cloud top
372 especially on the dry condition. The increase of CCN corresponds to the decrease of liquid and ice CER on the
373 same dry/wet condition, but when the moisture increases, the liquid CER becomes slightly larger. While we

374 should notice that the CDNC on dry or wet condition during heavy rainfall is naturally different, with the
375 average value of 1614.2 cm^{-3} on dry condition and 2066.2 cm^{-3} on wet condition, which we cannot fix in an
376 observation study. That is to say, when we divided the rainfall samples just by CDNC, the polluted condition
377 with more CDNC actually stands for the situation of more CDNC and more moisture, and the clean condition
378 represents the situation of less CDNC and less moisture. Thus, the results in Sect. 5.1 actually reflect the
379 combined effect of CCN and moisture, which is consistent with the pure CCN effect mentioned above,
380 indicating that the aerosol effect on these cloud properties is dominant on the polluted days.

381

382 **6 Discussion**

383 **6.1 Possible effect of aerosols on cloud with inclusion of moisture**

384 We attempt to understand the above results of aerosol effect on clouds with inclusion of moisture. The
385 aerosols serving as CCN can nucleate a larger number of cloud droplets and accumulate more liquid water in
386 the cloud, so the CF, COT and CWP become increased when the CCN increases or the moisture supply
387 increases as in Tab 3. However, why the effects of CCN and moisture on cloud top height are opposite have
388 not been clarified yet. Table 3 shows that the moisture could lift the cloud top height, which might due to the
389 increase of cloud water that causes the non-precipitating clouds growing to be higher. While for the result of
390 the lower cloud top height when CCN increases, we speculate it is because the precipitation process has
391 started thus the clouds could not grow to be higher since the rainfall start time is advanced in Fig 2b.

392 The decrease of liquid CER caused by CDNC in the same dry/wet condition (Tab. 3) can be interpreted by
393 Twomey effect that aerosols serving as CCN nucleate larger number concentrations of cloud drops, lead to the
394 decrease of cloud droplet size for competing the cloud water within a constant liquid water path (Squires and
395 Twomey, 1966; Twomey, 1977). When the moisture supply is more abundant, the liquid CER on the polluted
396 condition (group 4 in Tab. 3) is relatively increased compared with drier condition (group 2 in Tab. 3). This
397 might because the aerosols (CCN) increase the cloud droplet number, and the cloud water accordingly
398 increases with increased moisture supply, thus the cloud drops potentially become larger via the adequate
399 absorption of cloud water. We further investigate the relationship among CCN, CER and cloud water to verify
400 above hypothesis, shown as in Fig. 7. That is, the liquid CER exhibits significantly decreased along with the
401 increase of CDNC when fixing the cloud water. However, when increasing the cloud water, the liquid CER
402 becomes larger at the same value of CDNC.

403 The study also has shown the ice CWP increases and the ice CER decreases under pollution, and the ice
404 CER under pollution is still decreased when the moisture increases (Tab. 3). We assume the aerosols increase
405 the cloud droplets so that reduce the vapor pressure inside clouds, thus decrease the supersaturation and
406 weaken the process of transitions from liquid droplet into ice crystal, which is known as Bergeron process
407 (Squires, 1952). Currently the detailed physical processes of cold clouds and mixed-phase clouds are not clear,

408 including the diffusional grow, accretion, riming and melting process of ice precipitation (Cheng et al., 2010),
409 which needs numerical model simulations to be further explored.

410

411 **6.2 Different roles of aerosol radiative effect and cloud effect in heavy rainfall**

412 In Sect. 3 we found that the heavy rainfall has earlier start time and peak time, and longer duration on the
413 polluted condition. And afterwards, the earlier start of rainfall under pollution was found related to absorbing
414 aerosols mainly referring to BC (Fig. 3a&5a). We also compared the effect of BC on the associated clouds.
415 Figure 8a shows the CF larger than 90% rarely occurs in high BC environment, which might be associated
416 with the semi-direct effect of BC (IPCC, 2013). This result indicates the influence of BC on the heavy rainfall
417 in Fig. 5a is mainly due to the radiative effect rather than the cloud effect. The mechanism of BC effect on the
418 heavy rainfall can be interpreted by our previous study (Zhou et al., 2018) as: BC absorbs shortwave radiation
419 during the daytime and warms the lower troposphere at around 850 hPa, and then increases the instability of
420 the lower to middle atmosphere (850-500hPa) so that enhances the local upward motion and moisture
421 convergence. As a result, the BC-induced thermodynamic instability of the atmosphere triggers the occurrence
422 of heavy rainfall in advance. Thus, the low-level heating effect of BC should play a dominant role in the
423 beginning of rainfall especially before the formation of clouds during the daytime.

424 The delayed start of heavy rainfall with higher scattering aerosols in Fig. 2a and higher sulfate in Fig. 4b is
425 consistent with many studies that both the radiative effect and cloud effect of sulfate-like aerosols could delay
426 or suppress the occurrence of rainfall (Guo et al., 2013; Wang et al., 2016; Rosenfeld et al. 2014). Sulfate-like
427 aerosols as scattering aerosols could prevent the shortwave radiation from arriving at the surface thus cool the
428 surface and stabilize the atmosphere, which suppresses the rainfall formation (Guo et al., 2013; Wang et al.,
429 2016). Sulfate-like aerosols serving as CCN can also suppress the rainfall by cloud effect through reducing the
430 cloud droplet size and thus suppressing the collision-coalescence process of cloud droplets (Albrecht 1989;
431 Rosenfeld et al. 2014). Figure 8b does shows that in contrast with BC, the CF larger than 90% is significantly
432 increased in the high sulfate environment, which indicates the sulfate-like aerosols have evident influence on
433 the clouds. We also verified that the cloud droplet shifts to a smaller size when the CDNC increases (Fig. 6) in
434 Sect. 5, indicating that the cloud effect of aerosols could lead to the delay of the heavy rainfall occurrence.
435 Another significant feature is the longer duration of heavy rainfall in the high scattering aerosol cases and high
436 sulfate cases (Fig 3c&5b). We speculate that the longer duration is caused by the cloud effect of sulfate-like
437 aerosols. When CCN increases over BTH region, the cloud droplet size is decreased but the cloud water is
438 increased (Fig. 6). Therefore, the rainfall start time is delayed for the reduced collision-coalescence of cloud
439 droplets, while the duration might be prolonged due to the significant increase of cloud water. To further
440 investigate the mechanism of longer duration, we need the assistance of numerical model simulations in the
441 future work.

442 Accordingly, we speculate that the earlier start time of heavy rainfall related to absorbing aerosols (BC) is
443 due to the radiative heating effect of absorbing aerosols, while the longer rainfall duration associated with the
444 scattering aerosols (sulfate) is mainly caused by the cloud effect of sulfate-like aerosols. As a summary using
445 a schematic diagram (Fig. 9) to illustrate how aerosols modify the heavy rainfall over BTH region. On one
446 hand, BC heats the lower troposphere, changing the thermodynamic condition of atmosphere, which increases
447 upward motion and accelerates the formation of cloud and rainfall. On the other hand, the increased upward
448 motion transports more sulfate-like particles into the clouds so that more CCN and sufficient moisture
449 increase the cloud water, thus might prolong the duration of rainfall. As a result, the heavy rainfall shows
450 earlier start and peak time, and longer duration due to the combined effect of aerosol radiative effect and cloud
451 effect. To further distinguish the radiative effect and cloud effect of aerosols, we need to conduct numerical
452 model simulations in our future study.

453

454 **6.3 Uncertainties of different indicators**

455 In this study, we used two indicators to discriminate the different pollution levels, which are AOD and CDNC.
456 AOD is a good proxy for the large-scale pollution level, but it cannot well represent CCN (Shinozuka et al.,
457 2015). The value of AOD is influenced by moisture condition (Twohy et al., 2009). CDNC is a better proxy
458 for CCN, but it also has its uncertainties because it is calculated by the COT and CER. We can draw the same
459 conclusion on heavy rainfall diurnal changes between clean and polluted condition when using AOD and
460 CDNC respectively (Fig. 2). But when investigating the differences of cloud properties between clean and
461 polluted condition, there is a different result between using AOD and using CDNC, that the liquid CER is
462 decreased when CDNC increases (Fig. 6) while the liquid CER is increased when AOD increases. The
463 difference might be related with that the measurement biases, e.g., satellite AOD is evidently influenced by
464 the cloud (Brennan et al., 2005).

465 We applied ultraviolet AI and AOD of BC/sulfate to identify different types of aerosols. Ultraviolet AI in
466 this study is only used to detect the extreme circumstances of absorbing aerosols and scattering aerosols since
467 the near-zero values have larger uncertainties due to the cloud and other factors. The comparisons of
468 BC/sulfate AOD cases also have uncertainties because they are retrieved from MACC reanalysis data.
469 Although the four indicators all have their own uncertainties, we cannot find the more reliable datasets in a
470 long-term observational record, and the major findings can be well shown in these four indices.

471

472 **7. Conclusions**

473 Using the gauge-based hourly rainfall records, aerosol and cloud satellite products and high temporal
474 resolution reanalysis datasets during 2002-2012, this study investigated the different characteristics of heavy
475 rainfall in the diurnal time scale on the clean and polluted conditions respectively. Based on two indicators

476 that are AOD from MODIS aerosol product and retrieved CDNC from MODIS cloud product, we found three
477 features of rainfall changing by aerosols that the rainfall start and peak time occur earlier and the duration
478 becomes longer. The quantitative differences exist between the two indicators, i.e., the statistic differences of
479 above features between clean and polluted conditions are 0.7, 1.0, 0.8 hours based on AOD and 2.1, 4.2, 2.4
480 hours based on CDNC. The different roles of absorbing aerosols and scattering aerosols in modifying the
481 diurnal shift were also distinguishable using ultraviolet AI from OMI and reanalysis AOD of two aerosol types
482 (BC and sulfate). The absorbing aerosols (BC) correspond to the earlier start time and peak time of heavy
483 rainfall, while the scattering aerosols (sulfate) correspond to the delayed start time and the longer duration.

484 By comparing the characteristics of cloud macrophysics and microphysics variables, we found the CF, COT
485 (liquid and ice), CWP (liquid and ice) are increased on the polluted condition based on CDNC, but the cloud
486 top height and the CER (liquid and ice) are reduced. Considering moisture effect, the influence of aerosols on
487 COT and CWP is relatively larger than the moisture effect, although both aerosols and moisture could increase
488 the CF, COT and CWP. Liquid CER decreases almost a half under pollution, but when the moisture increases,
489 it shows a slight increase compared with the dryer condition. The influences of aerosols and moisture on cloud
490 top height are inverse, i.e., aerosols could lower the cloud top height while the moisture could lift the cloud
491 top.

492 According to these results, we speculate that both aerosol radiative effect and cloud effect have impacts on
493 the diurnal variation of heavy rainfall in BTH region. The heating effect of absorbing aerosols especially BC
494 increases the instability of the lower to middle atmosphere so that generates the heavy rainfall occurrence in
495 advance; and the increased aerosols nucleate more cloud droplets and accumulates more liquid water in clouds,
496 the duration of heavy rainfall is accordingly prolonged.

497 This study has clearly identified the aerosol effect on diurnal changes of heavy rainfall and concurrent
498 clouds in the BTH region and attempted to address the causes. However, although this work has attempted to
499 exclude the impacts from the meteorological background particularly circulation and moisture, the observation
500 study still has its limitation on studying aerosol effect on rainfall and cloud, such as the noise and uncertainty
501 of different observational data, the interaction of aerosol and meteorological factors and the mixing of
502 different types of aerosols. Numerical model simulations are necessarily applied to examine the speculation
503 we proposed here. And the specific processes of aerosols effect on the mix-phased cloud precipitation
504 formation also needs further exploration in our future study.

505

506 **Data availability**

507 We are grateful to the National Meteorological Information Centre (NMIC) of the China Meteorological
508 Administration (CMA) for providing hourly precipitation datasets. MODIS aerosol and cloud data were
509 obtained from <http://ladsweb.modaps.eosdis.nasa.gov>; ultraviolet AI data from OMI was obtained from

510 <https://daac.gsfc.nasa.gov/datasets?keywords=OMI&page=1>; MACC-II and ERA-interim reanalysis datasets
511 were obtained from <http://apps.ecmwf.int/datasets>.

512 **Author contributions**

513 JY and SZ conceived the study. SZ processed data and drew the figures. SZ and JY analyzed the observational
514 results and WCW, CZ and DG gave the professional guidance. PS provided the hourly precipitation dataset.
515 SZ and JY prepared the manuscript with contributions from WCW and CZ.

516 **Competing interests**

517 The authors declare that they have no conflict of interest.

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763 **Tables**

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Indicator	Data from	Time	Clean/less (< 25th)	Polluted/more (>75th)
AOD	MODIS	2002-2012	0.98	2.00
CDNC (cm ⁻³)	MODIS	2002-2012	699.20	2544.87
AAI	OMI	2005-2012	0.13	0.52
SAI	OMI	2005-2012	- 0.13	- 0.35
BC AOD	MACC	2003-2012	0.04	0.06
Sulfate AOD	MACC	2003-2012	0.46	0.87
Absolute humidity at 850 hPa (g/m ³)	ERA-interim	2002-2012	8.97	12.19

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767 Table 1. The indicators used in the study and their thresholds for clean/less and polluted/more conditions.

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Characteristics of heavy rainfall	Average of clean condition		Average of polluted condition		Difference (polluted - clean)		Significance of difference	
	AOD	CDNC	AOD	CDNC	AOD	CDNC	AOD	CDNC
Start time (LST)	24.2	24.5	23.5	22.4	- 0.7	- 2.1	P<0.05	P<0.05
Peak time (LST)	23.0	23.1	22.0	18.9	- 1.0	- 4.2	P<0.05	P<0.05
Duration (hours)	4.0	4.9	4.8	7.3	+ 0.8	+ 2.4	P<0.05	P<0.05
Intensity (0.1mm/hour)	164.9	167.0	169.6	162.0	+ 4.7	- 5.0	P>0.1	P>0.1

773

774 Table 2. The average values of start time (units: LST), peak time (units: LST), duration (units: hours) and
 775 intensity (units: 0.1mm/hour) of heavy rainfall respectively on clean condition and polluted condition using
 776 two indicators of AOD and CDNC, and the differences and significances of differences between clean and
 777 polluted conditions. “P<0.05” stands for the difference has passed the significance test of 95%, and “P>0.1”
 778 stands for the difference did not pass the significance test of 90%.

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Group (case number)	CF	CTP	COT (liquid)	COT (ice)	CWP (liquid)	CWP (ice)	CER (liquid)	CER (ice)
1.Clean, dry (140)	67.9	460.3	4.9	4.1	62.5	80.4	21.1	34.6
2.Polluted, dry (75)	73.1 <small>0.05<p_{1,2}<0.1</small>	541.1	21.3	25.4	154.3	432.4	11.6	29.2
3.Clean, wet (191)	85.8	405.1	6.3	7.4	79.3	150.8	20.3 <small>0.05<p_{1,3}<0.1</small>	34.7 <small>p_{1,3}>0.1</small>
4.Polluted, wet (338)	97.5	414.2 <small>p_{3,4}>0.1</small>	41.2	31.0 <small>0.05<p_{2,4}<0.1</small>	351.2	523.7 <small>p_{2,4}>0.1</small>	13.0	25.5

781

782 Table 3. The average values of CF (units: %), CTP (units: hPa), COT (liquid and ice, units: none), CWP
783 (liquid and ice, units: g/m²) and CER (liquid and ice, units: μm) in four groups. Grey numbers represent the
784 differences are not significant, in which “0.05<P<0.1” stands for the difference has passed the significance test
785 of 90% but did not pass the significance test of 95%, and “P>0.1” stands for the difference did not pass the
786 significance test of 90%.

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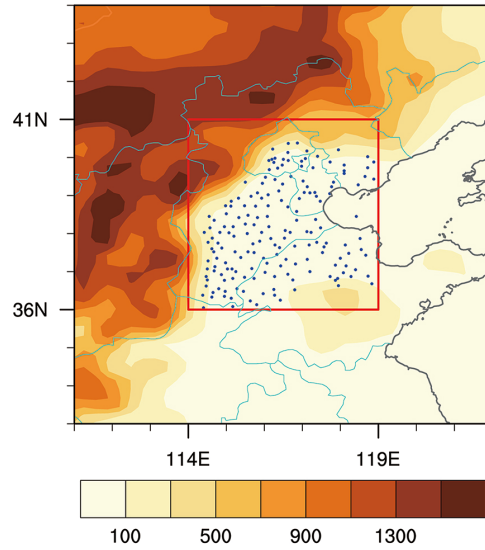
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808 **Figures**

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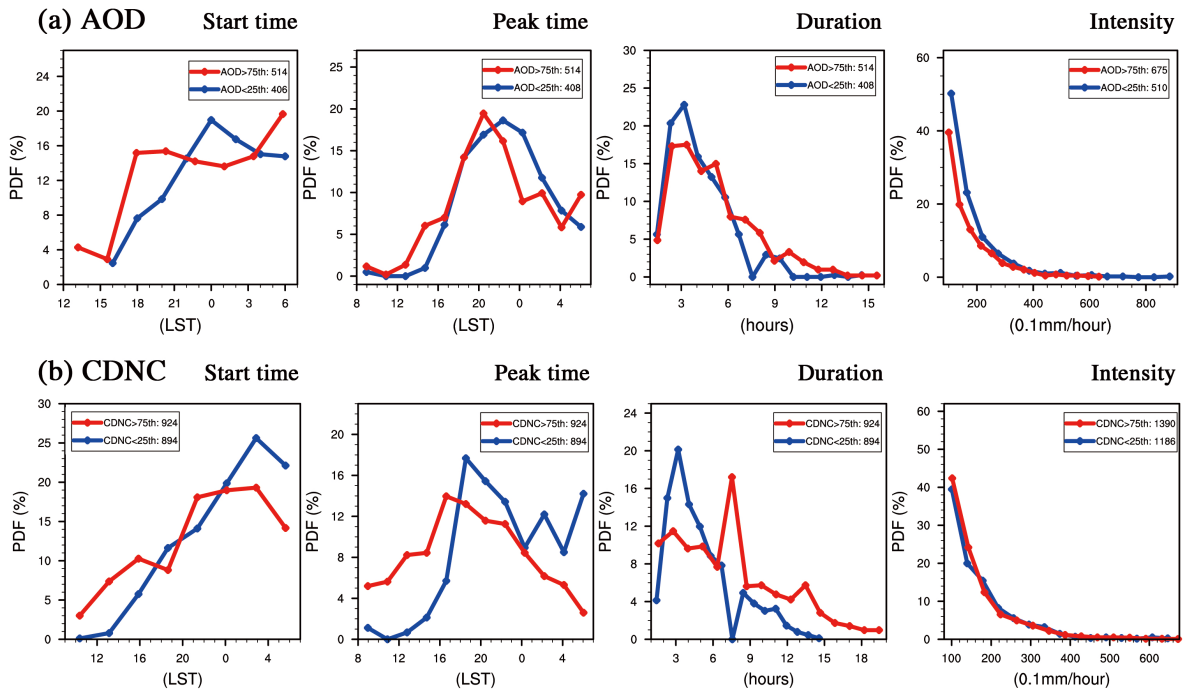


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811 Figure 1. Altitudes (shading, units: m) and selected stations (dots) in the BTH region (red box, 36–41° N,
812 114–119° E).

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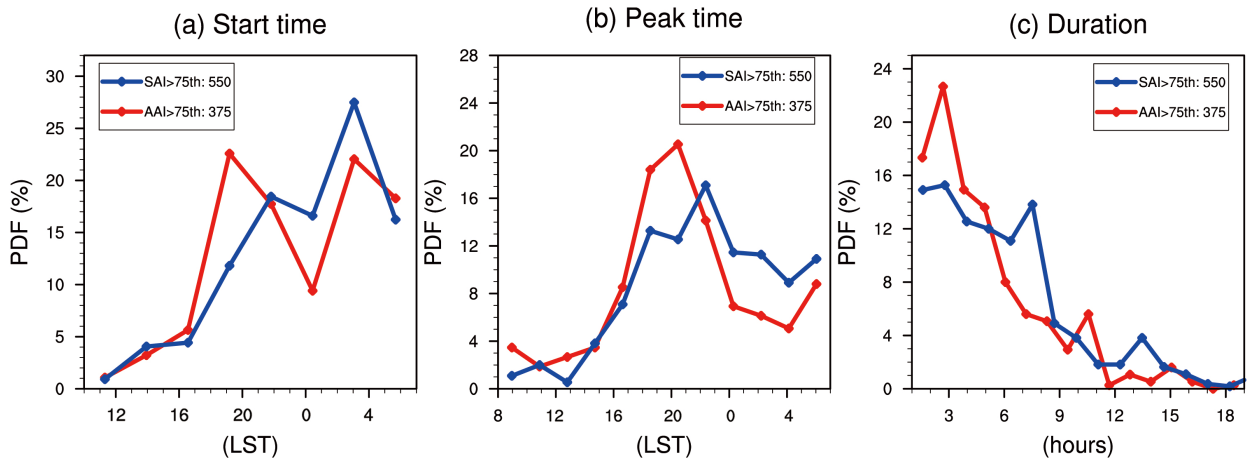


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816 Figure 2. PDF of start time (units: LST), peak time (units: LST), duration (units: hours) and intensity (units:

817 0.1mm/hour) of heavy rainfall on selected clean (blue lines) and polluted (red lines) conditions, respectively

818 using indicator of (a) AOD and (b) CDNC (cm^{-3}), during early summers from 2002 to 2012.



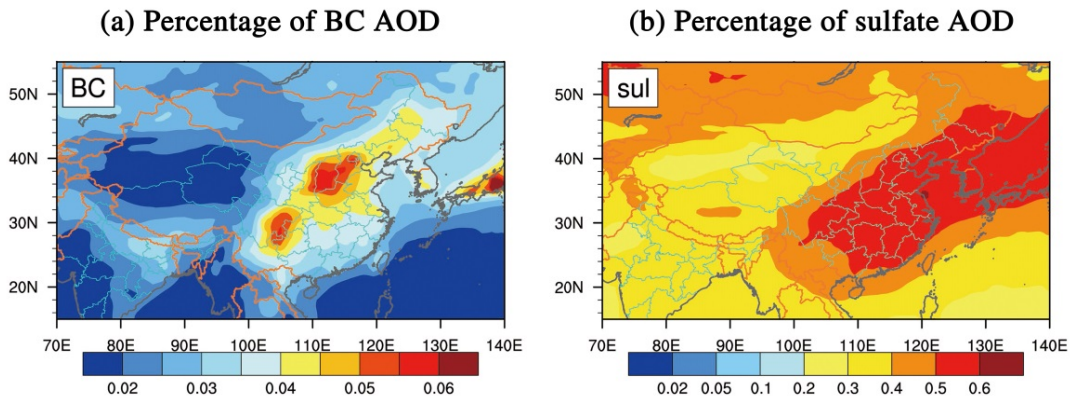
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821 Figure 3. PDF of (a) start time (units: LST), (b) peak time (units: LST), and (c) duration (units: hours) of
 822 heavy rainfall on the days that SAI more than 75th percentile (blue lines) and days that AAI more than 75th
 823 percentile (red lines), during early summers from 2005 to 2012. The differences between two groups have all
 824 passed the significant test of 95%.

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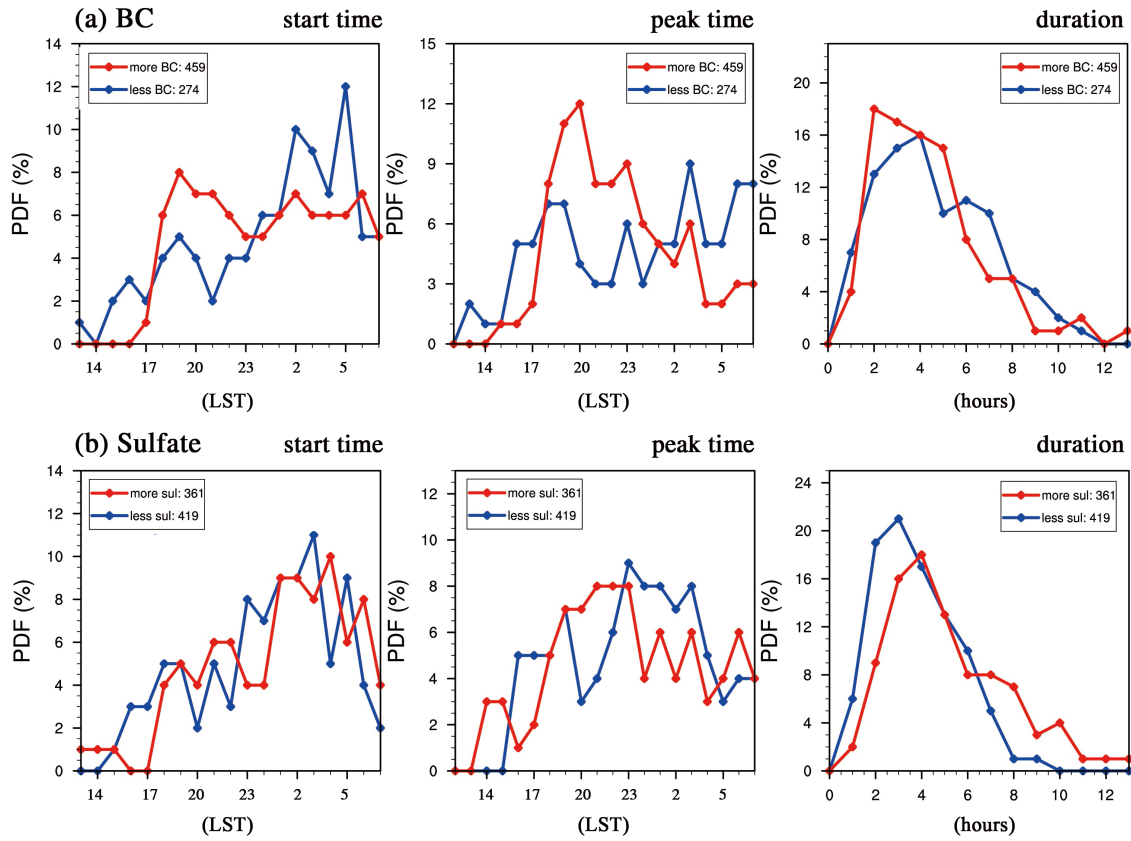


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829 Figure 4. Percentages of AOD for (a) BC and (b) sulfate in JJA during 2002 to 2012.

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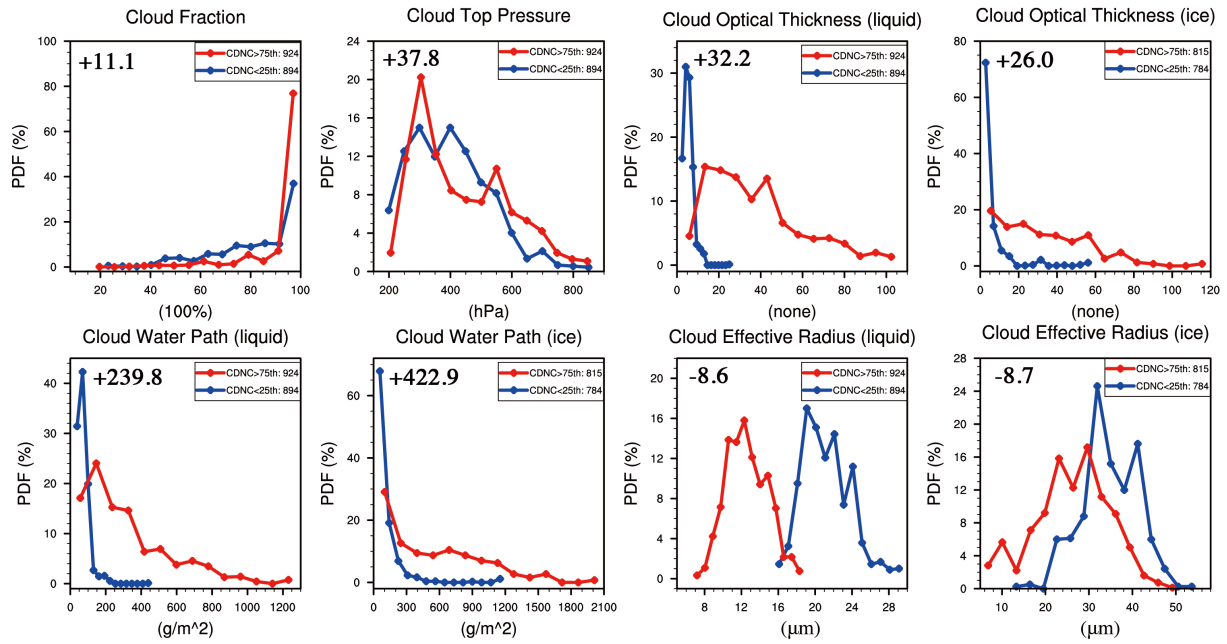
833 Figure 5. PDF of start time (units: LST), peak time (units: LST) and duration (units: hours) of heavy rainfall in
 834 different conditions of (a) BC and (b) sulfate. Blue/red lines stand for the condition of less/more BC or sulfate
 835 during early summers from 2003 to 2012. The differences have passed the significant test of 95%.

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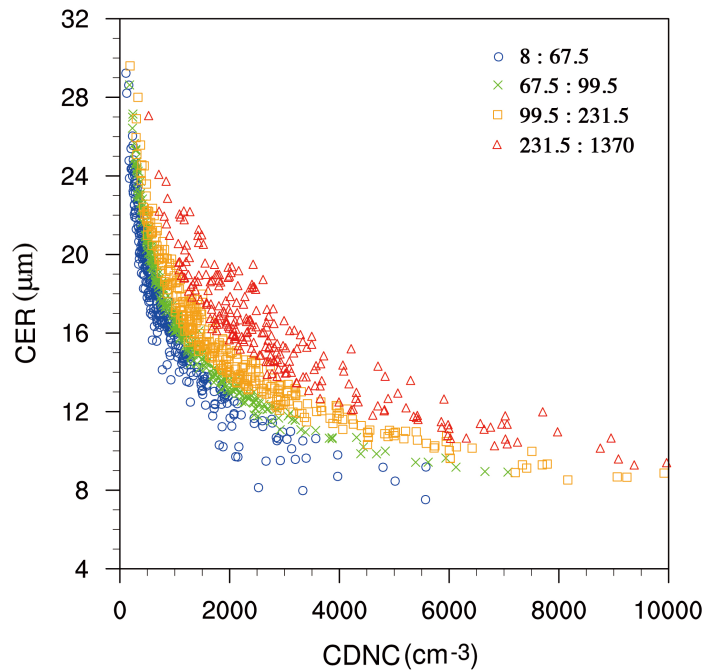
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841 Figure 6. PDF of CF (units: %), CTP (units: hPa), COT (liquid and ice, units: none), CWP (liquid and ice,
 842 units: g/m²) and CER (liquid and ice, units: μm) on selected clean (blue lines: CDNC < 25th percentile) and
 843 polluted (red lines: CDNC > 75th percentile) heavy rainfall days. The numbers in the upper left stand for the
 844 mean differences between polluted and clean days (polluted minus clean). The differences between clean and
 845 polluted cases have all passed the significant test of 95%.

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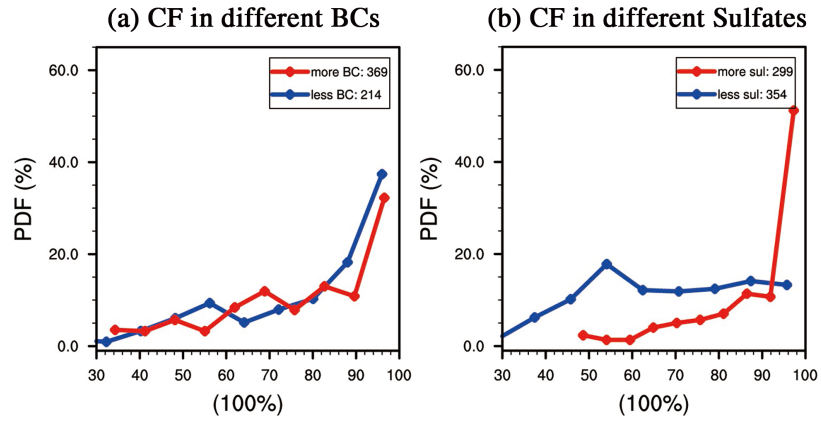
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848 Figure 7. Relationship of CER (units: μm) and CDNC (cm^{-3}) on different conditions of CWP (units: g/m²).

849 Different colors stand for different CWP conditions as shown in the legend.

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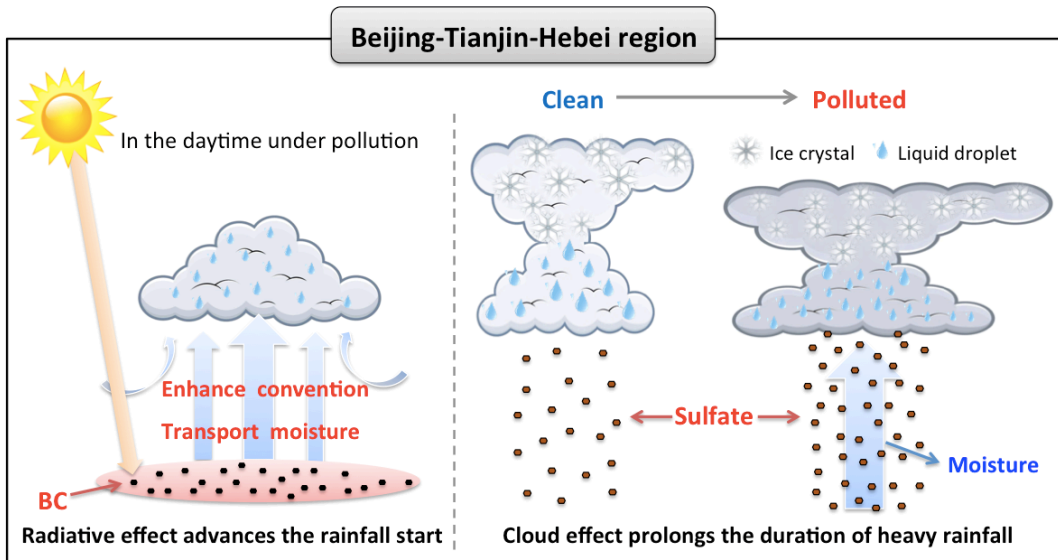
853 Figure 8. PDF of CF (units: 100%) respectively for selected less BC/sulfate (blue lines) and more BC/sulfate

854 (red lines) cases with heavy rainfall during 10 early summers (2003-2012).

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859 Figure 9. A schematic diagram for aerosols impact on heavy rainfall over Beijing-Tianjin-Hebei region.

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