Responds

To accommodate the comments from Reviewers and Editor, we have again made major revisions to the manuscript, as first summarized here:

1. We adjusted the structure of the article especially the "discussion" section to make it clearer and logical. The paper is re-organized as: The data and methodology are introduced in Sect. 2. Section 3 addresses the relationship between aerosol pollution and diurnal variation of heavy rainfall, covering the distinct characteristics of heavy rainfall using AOD and CDNC, the different behaviors of heavy rainfall along with different types of aerosols, and the comparison of heavy rainfall behaviors influenced respectively by moisture and aerosols. Section 4 describes the concurrent changes of cloud properties associated with aerosols and compares the possible influences of CDNC (CCN) and moisture on the cloud properties. Section 5 gives the hypothesis about the mechanisms of aerosol effects on the heavy rainfall. Conclusions and discussion are given in Sect. 6.

2. We have added additional results about the influence of moisture, such as the comparison in the influences of sulfate aerosols and moisture on the heavy rainfall and the results of rainfall characteristics when removing the high moisture samples in Sect. 3.3, as well as the results and discussions of cloud properties on different moisture conditions in Sect. 4.1&4.2. We also modified the contents concerning moisture in the abstract, conclusion and discussion.

3. We have added descriptions about the distribution of AOD and CDNC in Sect. 3, and other discussion that Reviewers raised, including the limitation of CDNC as surrogate of CCN concerning the updrafts and the uncertainties from the comparison between the extreme circumstances in Sect. 6.2.

More details are presented below:

Response P2:

A major issue of the manuscript remains that it is predicated on the assumption that the derived aerosol-cloud relationships are causal. Some caveats have been inserted in response to critical reviewer comments, e.g. related to relative humidity, but the overall wording of the manuscript remains fairly one-track and I agree with the reviewer that this appears to be more an afterthought. In a revised version of the manuscript, it is critical to separate the description of the derived relationships (e.g. for higher AOD the onset of precipitation occurs…) from a causal attribution (e.g. aerosols delay the onset of precipitation). Causal attribution needs to be presented and discussed in the context of confounding variables. Potential limitations of all causal attributions should be explicitly discussed. This critical analysis needs to be pulled through to the abstract and conclusions.

R: We have modified the whole structure and contents including the abstract and conclusions to make

it more comprehensive and objective as you mentioned, and added more results and discussions on the moisture influence in Sect. 3.3, 4.1 and 4.2. For example, we have tested the sensitivities of the changes of heavy rainfall and clouds associated aerosols to moisture in Sect.3.3&4.1, and compared and discussed the influences of sulfate/CCN and moisture on the rainfall and clouds in Sect. 3.3&4.2.

Response P3:

The use of CDNC as surrogate for CCN has severe limitations and it is not sufficient to simply declare the issue of updrafts out of scope of this study. Aerosol activation is controlled by CCN and updraft velocity. The region of interest is highly polluted so this is very likely to be an updraft limited regime (c.f. Reutter et al., ACP, 2009). Assuming this is the case, any presented correlation with CDNC with cloud properties could be interpreted as correlation of updraft velocity with the respective cloud property or precipitation – which are of course be expected, even under constant CCN. There are good reasons to believe that this could significantly contribute to or even dominate your presented correlations. If this analysis is retained, its reliability needs to be demonstrated. This cannot be ignored in the analysis and discussion.

R: We have added the discussion about the limitations of the use of CDNC as surrogate for CCN concerning the influence of the updraft (vertical velocity). We suppose that the CDNC could still represent the CCN for that: first, the results show that there is no significant correlation between CDNC and vertical velocity although the latter is larger in the polluted cases based on CDNC, and the percentage of the cases with CDNC more than 75th percentile and updraft more than 75th percentile in the cases only with updraft more than 75th percentile is only 30.6%. Second, the results of heavy rainfall characteristics based on CDNC do not change when limiting the vertical velocity to a certain range (less than 25th percentile, 25th – 75th percentile or more than 75th percentile). Third, we think the increase of vertical velocity on the polluted days is related to the increase of pollutants (Zhou et al., 2018), which means the pollutants including the aerosols serving as CCN and updraft are co-varied and they might be the cause and effect for each other. Therefore, we suppose the CDNC could stand for CCN to a certain extent although it might be partially dependent of updraft velocity, see Lines 592-604 Page 18.

Response P3:

You have now corrected your retrieved CDNC values by more than an order of magnitude from 2000cm-3 to a new value of 63cm-3, which seems unrealistically low for such a polluted region (c.f. Grosvenor et al., 2018). It appears neither of these values have been evaluated or critically analysed. This needs to be done thoroughly for this data to be used in the manuscript.

Given the significant structural uncertainties involved it is not appropriate to conclude on upper or lower bounds of the total effect from a single modelling study. Even if structural errors were negligible there exist a large number of parametric uncertainties that have not been sampled. The only dimension you sample is the perturbation strength so this need so be clear. However, given the extreme range of values chosen, these *perturbations are likely to act as on/off switch for some processes, such as autoconversion. The implications should be discussed.*

R: We have carefully double checked the calculation of CDNC and compared our values with other studies. The averaged CDNCs on the non-rainfall days, rainfall days and heavy rainfall days during 2002-2012 early summers are 54.70, 72.92, and 68.66 cm^{-3} respectively according to our calculation, which are similar with the global mean value 71.0 ± 26.4 cm^{-3} based on MODIS C6 product using 3 months spanning JJA, 2008 (Grosvenor et al., 2018). Although our values seem a little smaller for this polluted region, we think the distribution and the range (around 20-500 cm^{-3}) of the CDNC is reasonable when we look at the spectral distribution of CDNC (Fig. 2 in the manuscript). We admit there might be systematic biases in the calculation of CDNC in our study, but we think the values are acceptable after the check of the calculation and the comparisons with other studies (e.g. Zhu et al., 2018). We have added the description about the distributions and ranges of AOD and CDNC in Sect. 3, see Lines 251-264 Page 8.

It makes sense that comparing the extreme conditions of AOD and CDNC could bring uncertainties that these extreme conditions might be related with totally different microphysical process or meteorological background. Hence, to reduce the uncertainty, we further checked the results of heavy rainfall using the middle range of AOD and CDNC such as 25th – 50th percentile versus 50th -75th percentile. The results are basically the same and significant except the change of the peak time of heavy rainfall is not significant based on AOD. We added discussion about it, see Lines 625-632 Page 19.

Figures & Tables
All figures and tables need to be fully self-explaining and captions need to state the used data sources.

R: Thanks, modified and added.

References:

Gryspeerdt, E., Sourdeval, O., Quaas, J., Delanoë, J., Krämer, M., and Kühne, P.: Ice crystal number concentration estimates from lidar–radar satellite remote sensing – Part 2: Controls on the ice crystal number concentration, Atmos. Chem. Phys., 18, 14351–14370, doi:10.5194/acp-18-14351-2018, 2018.

Zhou, S., Yang, J., Wang, W. C., Gong, D., Shi, P., and Gao, M.: Shift of daily rainfall peaks over the Beijing–Tianjin–Hebei region: An indication of pollutant effects? Int. J. Climatol. 2018;1–10, doi:10.1002/joc.5700, 2018.

Zhu, Y., Rosenfeld, D., and Li, Z.: Under what conditions can we trust retrieved cloud drop concentrations in broken marine stratocumulus? J. Geophys. Res., 123, 8754-8767, doi:10.1029/2017JD028083, 2018.

An observational study of the effects of aerosols on diurnal variation of heavy rainfall and associated clouds over Beijing-Tianjin-Hebei

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Submitted to ACP

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Abstract: Our previous study found that the observed rainfall diurnal variation over Beijing-Tianjin-Hebei shows distinct signature of the effects of pollutants. Here we used the hourly rainfall data together with satellite-based daily information of aerosols and clouds to further investigate changes in heavy rainfall and clouds associated with aerosol changes. Because of the strong coupling effects, we also examined the sensitivity of these changes to moisture (specific humidity) variations. For heavy rainfall, three distinguished characteristics are identified: *earlier start time*, *earlier peak time*, and *longer duration*; and the signals are robust using aerosol indicators based on both aerosol optical depth and cloud droplet number concentration. In-depth analysis reveals that the first two characteristics occur in the presence of (absorbing) black carbon aerosols and that the third is related to more (scattering) sulfate aerosols and sensitive to moisture abundance. Cloud changes are also evident, showing increases in cloud fraction, cloud top pressure, the liquid/ice cloud optical thickness and cloud water path, and decrease in ice cloud effective radius; and these changes are insensitive to moisture. Finally, the mechanisms for heavy rainfall characteristics are discussed and hypothesized.

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

1. Introduction

Aerosols modify the hydrologic cycle through direct radiative and indirect cloud adjustment effects (IPCC, 2013). The direct effect, through absorbing and scattering solar radiation, leads to heating in the atmosphere (e.g. Jacobson 2001; Lau et al. 2006) and cooling on the surface (Lelieveld and Heintzenberg 1992; Guo et al. 2013; Yang et al., 2018), causing changes in atmospheric vertical static stability and subsequently modulation of rainfall (e.g. Rosenfeld et al. 2008). On the other hand, water-soluble aerosols serving as cloud condensation nuclei (CCN) affect the warm-rain and cold-rain processes through influencing the cloud droplet size distributions, cloud top heights and other cloud properties (Jiang et al., 2002; Givati and Rosenfeld 2004; Chen et al., 2011; Lim and Hong 2012; Tao et al., 2012). For Beijing-Tianjin-Hebei (BTH) the significant increase in pollution in recent decades has raised issues concerning aerosol-radiation-cloud-precipitation interactions. While the impact of aerosols on light rainfall or warm-rain processes is in general agreement among studies for this region (e.g., Qian et al., 2009), the uncertainties of the effects on heavy convective rainfall are still large (Guo et al., 2014; Wang et al., 2016).

The clouds that can generate heavy convective rainfall in BTH region usually contain warm clouds, cold clouds and mixed-phase clouds (e.g. Guo et al., 2015). Because the aerosol-cloud interactions in different types of clouds are distinct (Gryspeerdt et al., 2014b), aerosol indirect effect during heavy rainfall is more complicated than its direct effect (Sassen et al., 1995; Sherwood, 2002; Jiang et al., 2008, Tao et al., 2012). For warm clouds, by serving as CCN that nucleates more cloud droplets, aerosols can increase cloud albedo so called albedo effect or Twomey effect (Twomey, 1977), lengthen the cloud lifetime so called lifetime effect

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(Albrecht, 1989), and enhance thin cloud thermal emissivity so called thermal emissivity effect (Garrett and Zhao, 2006). The above effects tend to increase the cloud microphysical stability and suppress warm-rain processes (Albrecht 1989; Rosenfeld et al. 2014). For cold clouds and mixed-phase clouds, many studies reported that the cloud liquid accumulated by aerosols is converted to ice hydrometeors above the freezing level, which invigorates deep convective clouds and intensifies heavy precipitation so called invigoration effect (Rosenfeld and Woodley, 2000; Rosenfeld et al., 2008; Lee et al. 2009; Guo et al. 2014). The Twomey effect infers that aerosols serving as CCN that increase the cloud droplets could reduce cloud droplet size within a constant liquid water path (Twomey, 1977). However, the opposite results of relationship between aerosols and cloud droplet effective radius were reported in observations (Yuan et al., 2008; Panicker et al., 2010; Jung et al., 2013; Harikishan et al., 2016; Qiu et al., 2017), which might be related with the moisture supply near the cloud base (Yuan et al., 2008; Qiu et al., 2017). Besides, the influence of aerosols on ice clouds also depends upon the amount of moisture supply (Jiang et al., 2008). Therefore, how the aerosols modify the heavy convective rainfall and associated cloud changes does not reach a consensus, particularly if considering the different moisture conditions.

Heavy convective rainfall over BTH region usually occurs within a few hours, thus studying on the relationship between aerosols and rainfall diurnal variation could deepen our understanding of aerosol effects on heavy rainfall. Several previous studies have found that aerosols are related to the changes of the rainfall diurnal variation in other regions (Kim et al., 2010; Gryspeerdt et al., 2014b; Fan et al., 2015; Guo et al., 2016; Lee et al., 2016). However, the above studies do not address the change of cloud properties and its sensitivity to different conditions of moisture supply. Although our recent work over BTH region (Zhou et al. 2018) attempted to remove the meteorological effect including circulation and moisture and found that the peak of heavy rainfall shifts earlier on the polluted condition, it only excluded the extreme moisture conditions and focused on aerosol radiative effect on the rainfall diurnal variation. Therefore, this study aims to deepen the previous study (Zhou et al., 2018) through investigating the following questions: (1) how do aerosols (including absorbing aerosols and scattering aerosols) modify the behaviors of the heavy rainfall diurnal variation (start time, peak time, duration and intensity)? And what is the role of moisture in them? (2) how do aerosols influence the associated cloud properties with inclusion of moisture? To solve above questions, we used aerosol optical depth (AOD) as a macro indicator of aerosol pollution and cloud droplet number concentration (CDNC) as a micro indicator of CCN served by aerosols respectively to compare the characteristics of heavy rainfall diurnal variation and cloud properties between clean and polluted conditions, and applied aerosol index (AI) to distinguish the associated different effects of absorbing aerosols and scattering aerosols. In addition, we used the specific humidity (SH) at 850 hPa as an indicator of moisture supply condition to investigate the possible effects of moisture on the rainfall and clouds and compared them with the effects of aerosols. The paper is organized as following: The data and methodology are introduced in Sect. 2. Section 3 addresses the relationship between aerosol pollution and diurnal variation of heavy rainfall, covering the distinct characteristics of heavy rainfall using AOD and CDNC; the different behaviors of heavy

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rainfall diurnal variation along with different types of aerosols, and the comparison of heavy rainfall behaviors influenced respectively by moisture and aerosols. Section 4 describes the concurrent changes of cloud properties associated with aerosols and compares the possible influences of CDNC (CCN) and moisture on the cloud properties. Section 5 gives the hypothesis about the mechanisms of aerosol effects on the heavy rainfall. Conclusions and discussion will be given in Sect. 6.

2. Approach

2.1 Data

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

2.1.1 Precipitation

To study the diurnal variation of heavy rainfall, the gauge-based hourly precipitation datasets are used, which were obtained from the National Meteorological Information Center (NMIC) of the China Meteorological Administration (CMA) (Yu et al., 2007) at 2420 stations in China from 1951 to 2012. The quality control made by CMA/NMIC includes the check for extreme values (the value exceeding the monthly maximum in daily precipitation was rejected), the internal consistency check (wiping off the erroneous records caused by incorrect units, reading, or coding) and spatial consistency check (comparing the time series of hourly precipitation with nearby stations) [Shen et al., 2010]. Here we chose 176 stations in the plain area of BTH region that are below the topography of 100 meter above sea level as shown in Fig.1, because we purposely removed the probable orographic influence on the rainfall diurnal variation, which is consistent with our previous work (Zhou et al., 2018). The record analyzed here is the period of 2002 to 2012. We selected heavy rainfall days when the hourly precipitation amount is more than 8.0 mm/hour (defined by *Atmospheric Sciences Thesaurus, 1994*). Here "a day" is counted from 8 LST to 8 LST next day (0 UTC to 24 UTC).

2.1.2 Aerosols

In this study, we used two satellite data and one reanalysis data to investigate the aerosol optical amount and distinguish the different aerosol types.

AOD is a proxy for the optical amount of aerosol particles in a column of the atmosphere and serves as the macro indicator for the division of aerosol pollution condition in this study, which was obtained from MODIS (Moderate Resolution Imaging Spectroradiometer) Collection 6 L3 aerosol product with the horizontal resolution of 1°x1° onboard the Terra satellite (Tao et al., 2015). The quality assurance of marginal or higher confidence is used in this study. The reported uncertainty in MODIS AOD data is on the order of (-0.02-10%), (+0.04+10%) (Levy et al., 2013). The Terra satellite overpass time at the equator is around 10:30 local solar time (LST) in the daytime, and the satellite data is almost missing when it is rainy during the overpass time.

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As shown in Fig.3, the occurrence of selected heavy rainfall events in this study is mainly later than the satellite overpass time. Therefore, the AOD used here represents the situation of the air quality in advance of heavy rainfall appearance. Many studies have indicated the value of AOD is influenced by moisture condition, which is aerosol humidification effect (Twohy et al., 2009; Altaratz et al., 2013). Hence, we comprehensively analyzed the moisture effect on the rainfall and tried to remove the moisture effect from the relationship between aerosols and rainfall/clouds.

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The ultraviolet AI from Ozone Monitoring Instrument (OMI) on board the Aura satellite which was launched in July 2004, is used for detecting the different types of aerosols in this study. The OMI ultraviolet AI is a method of detecting absorbing aerosols from satellite measurements in the near-ultraviolet wavelength region (Torres et al., 1998). The positive values of ultraviolet AI are attributed to the absorbing aerosols such as smoke and dust while the negative values of AI stand for the non-absorbing aerosols (scattering aerosols) such as sulfate and sea salt (Tariq and Ali, 2015). The near-zero values of AI occur when clouds and Rayleigh scattering dominate (Hammer et al., 2018). Considering the near-zero values have more uncertainties, we only compare the extreme circumstances of absorbing aerosols and scattering aerosols in this study. The horizontal resolution of AI data is 1°×1° and it covers the period of 2005 to 2012.

MACC-II (Monitoring Atmospheric Composition and Climate Interim Implementation) reanalysis product produced by ECMWF (the European Centre for Medium-Range Weather Forecasts), provided the AOD datasets for different kinds of aerosols (BC, sulfate, organic matter, mineral dust and sea salt). MACC-II reanalysis products are observationally-based within a model framework, which can offer a more complete temporal and spatial coverage than observation and reduce the shortcomings of simulation that fail in simulating the complexity of real aerosol distributions (Benedetti *et al*., 2009). The horizontal resolution of MACC-II is also 1°×1° with the time interval of six-hour, and the results in the analysis of heavy rainfall show consistent based on the daily mean values which is shown in the figures and morning values that before the occurrence of heavy rainfall. MACC-II data covers the period of 2003 to 2012.

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2.1.3 Clouds

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Daily cloud variables, including cloud fraction (CF), cloud top pressure (CTP), cloud optical thickness (COT, liquid and ice), cloud water path (CWP, liquid and ice) and cloud effective radius (CER, liquid and ice), were obtained from MODIS Collection 6 L3 cloud product onboard the Terra satellite. The MODIS cloud product combines infrared emission and solar reflectance techniques to determine both physical and radiative cloud properties (Platnick et al., 2017). The validation of cloud top properties in this product has been conducted through comparisons with CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) data and other lidar observations (Holz et al., 2008; Menzel et al., 2008), and the validation and quality control of cloud optical products is performed primarily using in situ measurements obtained during field campaigns as well as the MODIS Airborne Simulator instrument (https://modis-atmos.gsfc.nasa.gov/products/cloud). Consistent with AOD, the measure of above cloud variables is before the occurrence of heavy rainfall.

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

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

Where C_w is the moist adiabatic condensate coefficient, and its value depends slightly on the temperature of the cloud layer, ranging from 1 to 2. 5 x 10^{-3} gm^{-4} for a temperature between 0 ℃ and 40 ℃ (Brenguier, 1991). In this study, we calculated the C_w through the function of the temperature (see Fig.1 in Zhu et al., 2018) at a given pressure that is 850 hPa. And we have tested the sensitivity of CDNC to the amount of C_w and found it almost keeps the same when the C_w changes from 1 to 2. 5 x 10^{-3} gm^{-4}. The coefficient k is the ratio between the volume mean radius and the effective radius, and varies between 0.5 and 1 (Brenguier et al., 2000). Here we used k = 1 for that we cannot get the accurate value of k and the value of k does not influence the rank of CDNC for the division of aerosol condition in this study. ρ_w is cloud water density. τ and Re are the liquid COT and CER obtained from MODIS Collection 6 L3 cloud product with resolution of 1°x1°. To reduce the uncertainty of CDNC retrieval caused by the heterogeneity effect from thin clouds (Nakajima and King, 1990; Quaas et al., 2008; Grandey and Stier, 2010; Grosvenor et al., 2018), we selected the CF more than 80%, the liquid COT more than 4 and the liquid CER more than 4 μm when calculating the CDNC (Quaas et al., 2008).

2.1.4 Other meteorological data

In this study, wind, temperature, pressure and SH data, were obtained from the ERA-Interim reanalysis datasets with 1°x1°horizontal resolution and 37 vertical levels at six-hour intervals. The daily mean values of these variables are used in the study, and we also verified the results based on the morning values that before the occurrence of heavy rainfall. ERA-Interim is a global atmospheric reanalysis produced by ECMWF, which covers the period from 1979 to near-real time (Dee et al., 2011). The SH, which stands for the water vapor content, serves as the indicator of moisture supply condition in this study.

2.2 Methodology

We used both station data of gauge-based precipitation and gridded data including aerosols, clouds and other meteorological variables. Gridded datasets in this study were downloaded with the horizontal resolution of 1°×1°, which are consistent with the resolution of MODIS L3 products. To unify the datasets, we interpolated all the gridded datasets onto the selected 176 rainfall stations using the average value in a 1°×1° grid as the

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background condition of each rainfall station, i.e., the stations in the same 1°×1° grid have the same aerosol, cloud and meteorological conditions.

2.2.1 Selection of sub-season and circulation

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

2.2.2 Classification of clean/polluted cases and moisture conditions

With the circulation of southwesterly, we used two indicators to distinguish the clean and polluted conditions from macro and micro perspectives, which are AOD and CDNC. The 25th and 75th percentiles of AOD/CDNC of the whole rainfall days are used as the thresholds of clean and polluted conditions, and the values are shown in Tab.1. There are 514 cases of heavy rainfall on the polluted days and 406 cases of that on the clean days when using AOD, and 630/716 cases on the polluted/clean condition when using CDNC (Fig. 3).

The absorbing aerosols are detected using the positive values of AI that is named as absorbing aerosol index (AAI) here, and we can retrieve the scattering aerosol index (SAI) using the negative values of AI. AAI and SAI are also divided into two groups using the threshold of 25th/75th percentile as shown in Tab.1. We used AAI/SAI more than 75th percentile as the extreme circumstances of absorbing/scattering aerosols to compare their impacts on the heavy rainfall. The sample numbers are 375 and 550 respectively for the extreme AAI and SAI cases. Using the same method, we chose cases with more BC/sulfate when the AOD of BC/sulfate is larger than the 75th percentile of itself in all rainy days, and cases with less BC/sulfate when that is less than the 25th percentile of itself in the same situation. Accordingly, we selected 459 heavy rainfall cases with more BC and 274 cases with less BC. Similarly, 361 cases with more sulfate and 419 cases with less sulfate were selected (Fig. 6).

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

2.2.3 Statistical analysis

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

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between the different groups of aerosol conditions. The differences between any two groups that have passed 95% statistical confidence level are considered significant. And two variables are considered correlated when the correlation coefficient is more than 0.5 or less than -0.5.

3. Changes of heavy rainfall

In this study, we used two indicators (AOD and CDNC) to identify the aerosol pollution. AOD is usually used as the macro indicator of aerosol pollution, which represents the optical amount of aerosol particles. However, AOD is not a proper proxy for CCN (Shinozuka et al., 2015), but the property of aerosols serving as CCN should be considered because aerosol-cloud interaction plays an indispensable role on changing rainfall diurnal variation. Therefore, here we applied the retrieved CDNC as the indicator of CCN (Zeng et al., 2014; Zhu et al., 2018).

We first investigated the value distribution of AOD and CDNC over the BTH region. Figure 2a&b shows the PDFs of AOD and CDNC on the non-rainfall days, rainfall days and heavy rainfall days respectively. The spectral distributions of AOD on different conditions are quite similar that the ranges are all between 0-5 and the peaks occur at around 1.2 (Fig. 2a). In contrast, CDNC shows different ranges between different conditions, that it ranges from around 20 cm^{-3} to 500 cm^{-3} on the rainfall days and non-rainfall days while from around 30 cm^{-3} to 420 cm^{-3} on the heavy rainfall days. Besides, the proportion of low CDNC is quite high on the non-rainfall days (Fig. 2b). The averaged CDNCs on the non-rainfall days, rainfall days and heavy rainfall days are 54.70, 72.92, and 68.66 cm^{-3} respectively. According to the above results, the range of AOD remains similar on the heavy rainfall days while the range of CDNC is shortened, probably because the cloud droplets become larger before heavy rainfall so that the number concentration becomes less. Therefore, to obtain comparable samples, we use percentile method to select respective clean and polluted cases based on above two indicators and compare the characteristics of heavy rainfall. Hence the heavier pollution means larger optical amount of aerosols measured by AOD, and more aerosols that could serve as CCN measured by CDNC.

3.1 Characteristics

Our previous study (Zhou et al. 2018) has reported the distinct peak shifts of rainfall diurnal variation between clean and polluted days using the indicator of AOD over the BTH region during early summer. Similar with our previous study, the PDF of the heavy rainfall peak time shows that the maximum of rainfall peak is about two hours earlier on the polluted days (20:00 LST) than that on the clean days (22:00 LST) (Fig. 3a). To comprehensively recognize the changes of rainfall diurnal variation associated with air qualities, here we examined the PDF of the start time, the duration and the intensity besides the peak time of heavy rainfall.

As shown in Fig. 3a, the start time of heavy rainfall exhibits a significant advance on the polluted days. The secondary peak on the early morning is ignored here because the early-morning rainfall is usually associated

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with the mountain winds (Wolyn et al., 1994; Li et al., 2016) and the nighttime low-level jet (Higgins et al., 1997; Liu et al., 2012) that is beyond the scope of this study. The time for the maximum frequency of heavy rainfall initiation is around 6 hours earlier on the polluted days, shifting from around 0:00 LST on the clean days to the 18:00 LST (Fig. 3a). Regarding the rainfall durations, the average persistence of heavy rainfall on polluted days is 0.8 hours longer than that on clean days (Tab. 2). According to the PDF shown as in Fig. 3a, the occurrence of short-term precipitation (≤6 hours, Yuan et al., 2010) decreases while that of long-term precipitation (>6 hours, Yuan et al., 2010) increases. The intensity of hourly rainfall exhibits a non-significant increase on the polluted days.

The distinct behaviors of heavy rainfall diurnal variation between clean and polluted days have been well demonstrated using the indicator of AOD. Using CDNC as the indicator of CCN, the above-mentioned results are also significant, as shown in Fig. 3b. The start time and peak time of heavy rainfall on the polluted condition also show significant advances compared with that on the clean condition, with the average advances of 1.4 hours and 3.0 hours respectively (Tab. 2). The duration of heavy rainfall on the polluted condition is also prolonged, which is 2.2 hours longer in average (Tab. 2). Similar with the results based on AOD, the difference of rainfall intensity between clean and polluted conditions using CDNC does not pass the 95% statistical confidence level as well.

Hence, the results using either AOD or CDNC show that the start and peak time of heavy rainfall occur earlier and the duration becomes longer under pollution, although there are some quantitative differences between the two indicators. We found the AOD and CDNC only have a non-significant positive correlation, which denotes that the selected cases could be different between using AOD and CDNC. The cases of heavy rainfall using CDNC seem more extreme, because the rainfall behaviors exhibit more evident changes using CDNC than using AOD. The result differences between the two indicators might be attributed to the non-linear relationship between CCN and aerosol pollution (e.g., Jiang et al., 2016), the misdetection of AOD when the humidity is high (Boucher and Quaas, 2012), the calculation uncertainty of CDNC, and the sampling differences between AOD and CDNC. Since the two indicators represent aerosols from the different perspectives, we cannot identify which one is more reliable. Because the change of rainfall intensity is not significant using either AOD or CDNC, the following analysis only focuses on studying the start time, peak time and duration of heavy rainfall along with aerosol pollution.

3.2 Sensitivities to aerosol types

Using the indicator of AI, we further investigated the distinct behaviors of heavy rainfall diurnal variation related to absorbing aerosols and scattering aerosols respectively. The PDF of start time, peak time and duration of heavy rainfall under the extreme circumstances of absorbing aerosols and scattering aerosols are compared in Fig. 4. Here, we briefly named the days with extreme large amount of absorbing aerosols as absorbing aerosol days and with more scattering aerosols as scattering aerosol days. The start time of heavy rainfall on absorbing aerosol days shows a significant earlier compared with that on scattering aerosol days

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(Fig. 4a), with 0.7 hours advance in average (Tab. 3). Similarly, the rainfall peak time also shows earlier on absorbing aerosol days (Fig. 4b), with an average advance of 1.6 hours (Tab. 3). The rainfall duration on scattering aerosol days shows longer than that on absorbing aerosol days, which are 6.0 hours and 5.0 hours respectively in average (Tab. 3). All the above-mentioned differences between the two groups have passed 95% statistical confidence level. The results indicate that the absorbing aerosols and scattering aerosols may have different or inverse effects on the heavy rainfall that absorbing aerosols may generate the heavy rainfall in advance while the scattering aerosols may delay and prolong the heavy rainfall.

To further verify the different behaviors of heavy rainfall diurnal variation associated with two different types of aerosols, we purposely re-examine the above-mentioned phenomena using BC/sulfate that can represent typical absorbing/scattering aerosols over the BTH region. BC has its maximum center over BTH region (Fig. 5a) and our previous study has indicated that the radiative effect of BC low-level warming may facilitate the convective rainfall generation (Zhou et al., 2018). The percentage of sulfate is also large over the BTH region (Fig. 5b) and the sulfate is one of the most effective CCN that influences the precipitation in this region (Gunthe et al., 2011). Accordingly, we selected the cases with different amounts of BC and sulfate AOD to compare their roles on the diurnal variation of heavy rainfall. The methods have been described in Sect. 2.2.2. The PDF of the start time, peak time and duration of heavy rainfall in the cases with more/less amount of BC are shown in Fig. 6a, respectively. The most striking result is that the maximum frequency of rainfall start time in the more BC cases evidently shifts earlier (Fig. 6a). Meanwhile, the mean peak time in the more BC cases shows 1.1 hour earlier than that in the less BC cases (Tab. 3). And the duration of heavy rainfall is slightly shortened by the averaged 0.2 hours in the more BC cases. The features in more BC cases are consistent with the above results of absorbing aerosols. In contrast, when the sulfate has higher amount, the mean start time of rainfall is delayed by 0.5 hours, while the duration shows a significant increase by 1.5 hours in average (Tab. 3). The behaviors in the more sulfate cases also exhibit similar with the above results of scattering aerosols, except for the peak time that shows later in the scattering aerosol cases but a little earlier in the more sulfate cases (Tab. 3).

3.3 Influence of moisture

Moisture supply is an indispensable factor for the precipitation formation, and it also has an important impact on AOD (Boucher and Quaas, 2012). Since the southwesterly circulation can not only transport pollutants but also plenty of moisture to the BTH region (Wu et al., 2017), more pollution usually corresponds to more moisture for the BTH region (Sun et al., 2015) so that it is hard to completely remove the moisture effect on the above results in the pure observational study. Here we attempt to recognize the moisture effect on the heavy rainfall to further understand the above aerosol-associated changes. Because the moisture supply for BTH is mainly transported via low-level southwesterly circulation, we purposely used the SH at 850 hPa as the indicator of moisture condition.

Using the similar percentile method with polluted/clean days, we compared the heavy rainfall

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characteristics in the more humid (more than 75th percentile) and the less humid (less than 25th percentile) environments regardless of the aerosol condition, as shown in Fig. 7a. The results show that the start time of heavy rainfall is delayed by 0.9 hours, the peak time is 0.6 hours earlier and the duration is prolonged by 2.0 hours in average in the more humid environment, which is similar with the results of the more sulfate cases. Besides, the same results are obtained using different moisture indicator, e.g. the 850 hPa absolute humidity. These results indicate the advance of heavy rainfall start time on the polluted days is not caused by more moisture supply, while the longer duration and earlier peak in the more sulfate cases might be related to the increased moisture supply. To further identify the role of sulfate, we tested the sensitivities of the results associated with sulfate when limiting the moisture condition. In the dry and intermediate cases (SH less than 25th percentile and SH between 25th -75th percentiles), the heavy rainfall still shows later start time, earlier peak and significant longer duration with the increase of sulfate, while the change of peak time is not significant in the dry cases and that of start time is not significant in the intermediate cases; in the high moisture cases (SH more than 75th percentile), it shows earlier peak and shorter duration in the more sulfate cases while the change of start time is not significant. Therefore, we suppose that the impact of sulfate aerosols on the heavy rainfall is sensitive to moisture, and notably the sulfate could contribute to the longer duration in the polluted cases when it is relatively dry.

We also investigate the distributions of moisture and rainfall behaviors in the clean and polluted cases respectively using AOD and CDNC (Fig. 7 b&c). The results show that the relationship between moisture and rainfall start time/peak time/duration is not linear. Using either AOD or CDNC, the distribution of SH exhibits a slight increase in the polluted cases, indicating that the polluted cases have the more moisture than the clean cases which is particularly well shown using AOD. However, when fixing the moisture at a certain range especially at the relative dry condition, we can detect the similar phenomena of earlier start/peak time and longer duration in the polluted cases. For example, when the amount of 850 hPa SH is between 8-12 g/kg, the start &peak time in the polluted cases show significant earlier and the duration exhibits slightly increased compared with that in the clean cases using either AOD or CDNC. To further clarify the characteristics of heavy rainfall associated with pollution, we removed the samples with high SH (SH more than 75th percentile) and found that the results in section 3.1 remain the same, that when SH is less than 12.95 g/kg (75th percentile), the start/peak time of heavy rainfall is also in advance and the duration is still prolonged with the increase of AOD/CDNC (Fig. 8).

The above results indicate that the advance of heavy rainfall start in the polluted cases is independent of moisture, while the advance of peak time and longer duration might be related to the moisture effect. For the peak time of heavy rainfall, although the results of BC, sulfate and moisture show consistent advance, we suppose the role of BC (absorbing aerosols) might be dominant compared with that of sulfate or moisture because the change of peak time in the former analysis is much larger (Tab. 3). Both sulfate and moisture may contribute to the longer duration of heavy rainfall (Fig. 6b&7a), but the role of sulfate seems sensitive to the moisture condition, that the duration in the more sulfate cases is longer when the moisture condition is

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relatively dry while becomes shorter when it is extremely wet. Because we cannot completely separate the sulfate and moisture, we are not quite clear about their individual roles at present. Overall, under the condition of removing the extremely high moisture cases, the earlier start/peak time and longer duration of heavy rainfall associated with aerosol pollution are significant and irrespective of moisture.

4. Changes of clouds

To understand the cloud effect of aerosols during heavy rainfall diurnal variation, we need to recognize the associated cloud characteristics on the clean and polluted conditions. The cloud properties we used were obtained from satellite product that were measured at the same time with aerosols before the occurrence of heavy rainfall. The differences of cloud features were examined in both macroscopic (including CF, CTP, COT and CWP) and microscopic properties (including CER) between the clean and polluted conditions based on AOD and CDNC respectively.

4.1 Characteristics

Using AOD as the macro aerosol indicator, as shown in Fig. 9, the PDF distribution of CF shows that the CF on the polluted condition is evidently larger than that on the clean condition. The average CF is 62.8% on the clean condition, and 89.3% on the polluted condition (Tab. 4), which is increased by 26.1%. The average CTP on the polluted condition is 487.3 hPa, which is larger than 442.3 hPa on the clean condition, indicating that the cloud top height is lower on the polluted days. The COT, CWP and CER were further analyzed for the liquid and ice portions of clouds as shown in Fig. 9. Both liquid and ice COT on the polluted condition exhibit significant increases compared with that on the clean condition. The mean amount of liquid COT is increased by 3.1 and ice COT increases by 6.2 (Tab. 4). Similar with COT, the amounts of liquid and ice CWP also increase under pollution, which increase by 33.6 g/m^2 and 88.2 g/m^2 respectively. In addition, the liquid CER is increased by 0.8 μm and the ice CER is decreased by 2.8 μm on the polluted days. The differences of above cloud properties between clean and polluted cases have all passed the 95% statistical confidence level.

Using CDNC as the micro aerosol indicator, the above-mentioned changes of cloud properties are consistent with that using AOD, except for liquid CER (Fig. 9). Since the calculation method of CDNC is not independent on the liquid COT and liquid CER, we would not directly compare the results of liquid COT and CER based on CDNC with those based on AOD here. But according to other variables that are independent of the CDNC calculation, we found the cases with more CDNC are accompanied with the increase of CTP, ice COT and liquid & ice CWP, which increase by 32.8 hPa, 24.4, 215.8 g/m^2 and 370.9 g/m^2 respectively (Tab 4) and all of which are consistent with the results based on AOD. The CER of ice clouds also shows a consistent decrease by 8.8 μm on the polluted condition based on CDNC. We noticed that the changes of COT/CWP/CER for both liquid and ice based on CDNC are much larger than that based on AOD, which indicates that these cloud properties might be more sensitive to the indicator of CDNC rather than AOD.

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According to the above comparison, the concurrent changes of cloud properties along with heavy rainfall diurnal variation show consistent results using the two aerosol indicators (AOD and CDNC). The pollution corresponds to the increase of CF, ice COT, liquid and ice CWP, but the decrease of cloud top height (the increase of CTP corresponds to the decrease of cloud top height) and ice CER. The liquid COT and liquid CER are also increased with the enhanced pollution in the AOD analysis. Besides, these above results exhibit significant when we limited the moisture to the dryer condition (SH less than 25^{th} percentile) or intermediate condition (SH more than 25^{th} percentile and less than 75^{th} percentile). When the moisture is higher (SH more than 75^{th} percentile), the change of CTP does not show significant based on CDNC.

For these results, we made the following speculation: First, the CF, liquid & ice COT and CWP increase with pollution, might because the aerosols serving as CCN can nucleate a larger number of cloud droplets and accumulate more liquid water in the cloud thus increase the CF, COT and CWP. Second, the CTP increases under pollution using both AOD and CDNC, which denotes the decrease of the cloud top height, might because the earlier start of the precipitation process (Fig. 3) inhibits the vertical growth of clouds. Third, the ice CER decreases under pollution using either AOD or CDNC, probably because the increased cloud droplet number leads to more cloud droplets transforming into ice crystals and causes the decrease of ice CER (Chylek et al., 2006; Zhao et al., 2018; Gryspeerdt et al., 2018). However, the results of liquid CER might have uncertainties. The liquid CER is increased when AOD increases (Fig. 9), which might be related to the aerosol humidification effect, the misdetection of AOD and cloud water, and also might result from the earlier formation of the clouds and heavy rainfall on the polluted days. Since we cannot distinguish the liquid part of mix-phased clouds from liquid (warm) clouds in the observation, the above changes of liquid cloud properties might come from both the liquid (warm) clouds and the liquid part of mixed-phase clouds. Likewise, the above-mentioned changes of ice cloud properties might come from both ice (cold) clouds and the ice part of mixed-phase clouds. Currently the detailed physical processes of cold clouds and mixed-phase clouds have been not clarified yet, including the diffusional grow, accretion, riming and melting process of ice precipitation (Cheng et al., 2010), which needs numerical model simulations to be further explored.

4.2 Sensitivities to CCN and moisture

Section 3.3 has shown that the diurnal variation of heavy rainfall with more moisture supply is similar with the changes of heavy rainfall with more sulfate aerosols. We assume that the moisture under the cloud base and the sulfate serving as CCN both influence the cloud properties (Yuan et al., 2008; Jiang et al., 2008; Jung et al., 2013; Qiu et al., 2017). To identify the effect of CCN on clouds and its sensitivity to moisture, using CDNC to represent CCN, we purposely investigated the changes of above cloud properties on the different conditions of the CDNC and the low-level moisture (850hPa SH) respectively.

We categorized all cases of heavy rainfall into four groups, which are (1) clean and dry, (2) polluted and dry, (3) clean and wet, (4) polluted and wet, and checked the changes of above cloud properties, as shown in Tab. 5. To retrieve the comparable samples, here "clean/polluted" refers to the CDNC on that day less/more

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than 25^{th}/75^{th} percentile of the CDNC among the heavy rainfall days, and similarly, the “dry/wet” refers to the SH on that day less/more than 25^{th}/75^{th} percentile of itself among the heavy rainfall days. The average CDNC is 68.58 cm^{-3} on the dry condition and 68.56 cm^{-3} on the wet condition, and the average SH is 11.3 g/kg and 11.8 g/kg on the clean and polluted conditions respectively, thus we can consider the CDNC or SH remains the same when the other condition changes. We made the significant test of differences between group 1 and 2, group 1 and 3, group 2 and 4, group 3 and 4. Because the CF is fixed above 80% when calculating the CDNC (see in Sect. 2.1.3), here the selected groups all belong to the condition of higher CF.

Comparing the results of group 1 and 2, which are both on the dry condition, we can identify the influence of CDNC on the cloud properties, which stands for the effect of CCN. The changes of these cloud variables are the same as that in Sect. 4.1, that the CF, ice COT and liquid & ice CWP are increased on the polluted condition, while the cloud top height and ice CER are decreased based on CDNC. Among these variables, the ice COT and liquid & ice CWP are especially larger on the polluted condition, which are 5-6 times larger than that on the clean condition (Tab. 5). On the wet condition, comparing the group 3 and 4, the changes are similar that the CF, ice COT and liquid & ice CWP are increased and the ice CER are decreased but the change of CTP becomes not significant. However, the changes of these variables on the dry condition are evidently enhanced than that on the wet condition, which indicates these cloud properties might be more sensitive to CDNC on the dry condition. The above comparisons indicate that with the increase of CDNC (CCN), the CF, ice COT and liquid & ice CWP are increased while the ice CER is decreased regardless of the moisture amount. Although the comparisons of liquid COT and liquid CER based on CDNC are meaningless since the CDNC is calculated by the two variables, we infer that the increase of liquid COT and the decrease of liquid CER (Tab. 5) might be not completely caused by CDNC calculation but the natural effect of CCN.

Comparing the results of group 1 and 3, we can get the changes of cloud properties related only to moisture on the same clean condition. A common feature is that CTP, COT and CWP both for liquid and ice exhibit increases along with the increase of moisture. Compared with the CTP on the clean and dry condition, it increases on both polluted & dry condition (group 2) and clean & wet condition (group 3), but on the former condition its increase is larger, which indicates the influence of moisture on CTP might be secondary compared to the CDNC (CCN) effect. Similarly, comparing the COT/CWP in group 2 and 3, the increases of COT and CWP both for liquid and ice in group 2 are 3-6 times larger than that in group 3, which indicates that the influences of moisture on COT and CWP may not overcome the influence of CCN. With the increase of moisture, the change of liquid CER is not significant on the same clean condition, but the ice CER is significantly decreased. On the polluted condition, comparing group 2 and 4, we found the COT and CWP both for liquid and ice on the wet condition are evidently smaller than that on the dry condition, which indicates that increasing the moisture might partly compensate for the influence of CDNC (CCN) on COT/CWP.

The results above indicate that both CDNC (CCN) and moisture have impacts on cloud properties. They

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both contribute to the increase of CF, COT and CWP, in which the influence of CDNC (CCN) on COT and CWP are significantly larger than moisture. The increase of either CDNC or moisture corresponds to the increase of CTP. But when the CDNC and moisture increase simultaneously, the CTP becomes smaller. Both CDNC and moisture correspond to the significant decrease of ice CER, while only CDNC corresponds to the decrease of liquid CER and that might be ascribed to the calculation method of CDNC. To reduce uncertainties, we have tested the SH at different levels (e.g., 700 hPa and 800 hPa) and different moisture indicator (e.g. absolute humidity) to verify these results, and found most cloud variables show the similar changes with above except for the CTP and the liquid CER, which indicates the changes of CTP and liquid CER are more sensitive and have larger uncertainties. Since the behaviors of cloud changes are similar along with the increase of either CDNC (CCN) or moisture but more sensitive to the former, the results in Sect. 4.1 might actually reflect the combined effect of CCN and moisture, and the aerosol effect on these cloud properties might be dominant on the polluted days.

Therefore, combining with the results in Sect. 3.3, although we cannot completely separate the aerosols and moisture, the CCN is assumed to play a vital role on the clouds and precipitation especially in a relatively dry environment. In the relatively wet environment, the CCN might have some inhibitory effect since the duration of heavy rainfall is shorter with the increase of sulfate when it is extremely wet, and the changes of cloud features along with the CDNC increase are smaller on the wet condition. Due to the limitations of observational study, we currently cannot figure out the respective roles of aerosols and moisture.

5. Hypothesis

According to all the above results, we have made hypotheses about the aerosol effects on the heavy rainfall over the BTH region. In Sect. 3.1 we found that the heavy rainfall has earlier start and peak time, and longer duration on the polluted condition. And afterwards, the earlier start of rainfall under pollution was found related to absorbing aerosols mainly referring to BC (Fig. 4a&6a). We also compared the effect of BC on the associated clouds. Figure 10a shows the CF larger than 90% rarely occurs in the more BC environment, which might be associated with the semi-direct effect of BC (Ackerman, 2000) or estimated inversion strength and BC co-vary. This result indicates the influence of BC on the heavy rainfall in Fig. 6a is mainly due to the radiative effect rather than the cloud effect. The mechanism of BC effect on the heavy rainfall can be interpreted by our previous study (Zhou et al., 2018) as: BC absorbs shortwave radiation during the daytime and warms the lower troposphere at around 850 hPa, and then increases the instability of the lower to middle atmosphere (850-500hPa) so that enhances the local upward motion and moisture convergence. As a result, the BC-induced thermodynamic instability of the atmosphere triggers the occurrence of heavy rainfall in advance. Thus, the low-level heating effect of BC might play a dominant role in the beginning of rainfall especially before the formation of clouds during the daytime.

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The delayed start of heavy rainfall with scattering aerosols in Fig. 4a and more sulfate in Fig. 6b is consistent with many studies that both the radiative effect and cloud effect of sulfate-like aerosols could delay or suppress the occurrence of rainfall (Guo et al., 2013; Wang et al., 2016; Rosenfeld et al. 2014). Sulfate-like aerosols as scattering aerosols could prevent the shortwave radiation from arriving at the surface thus cool the surface and stabilize the atmosphere, which suppresses the rainfall formation (Guo et al., 2013; Wang et al., 2016). Sulfate-like aerosols serving as CCN can also suppress the rainfall by cloud effect through reducing the cloud droplet size and thus suppressing the collision-coalescence process of cloud droplets (Albrecht 1989; Rosenfeld et al. 2014). Figure 10b does shows that in contrast with BC, the CF larger than 90% is significantly increased in the more sulfate environment, which indicates the sulfate-like aerosols might have more evident influence on the clouds and subsequently the rainfall changes associated with sulfate are probably due to the cloud effects. Another significant feature is the longer duration of heavy rainfall in the scattering aerosol cases, more sulfate cases and high moisture cases (Fig 4c, 6b&7a). We speculate that the postponed start of heavy rainfall is mainly due to the effect of sulfate-like aerosols, while the longer duration is caused by both the cloud effect of sulfate-like aerosols and the increased moisture supply, because increasing either CCN or the moisture supply can increase cloud water (Sect. 4.2), which could lead to the longer rainfall duration. To further investigate the mechanism of longer duration, we need the assistance of numerical model simulations in the future work.

Accordingly, we speculate that the earlier start time of heavy rainfall related to absorbing aerosols (BC) is due to the radiative heating of absorbing aerosols, while the longer rainfall duration is probably caused by both the cloud effect of sulfate-like aerosols and the increased moisture supply. As a summary we use a schematic diagram (Fig. 11) to illustrate how aerosols modify the heavy rainfall over the BTH region. On one hand, BC heats the lower troposphere, changing the thermodynamic condition of atmosphere, which increases the upward motion and accelerates the formation of clouds and rainfall. On the other hand, the increased upward motion transports more sulfate-like particles and moisture into the clouds so that more CCN and sufficient moisture increase the cloud water, thus might prolong the duration of rainfall. As a result, the heavy rainfall over BTH region in southwesterly shows earlier start and peak time, and longer duration might due to the combined effect of aerosol radiative effect, aerosol cloud effect as well as the moisture effect. To further distinguish the individual effect, we need to conduct numerical model simulations in our future study.

6. Conclusions and discussion

6.1 Conclusions

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

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aerosol indicators that are AOD from MODIS aerosol product and calculated CDNC from MODIS cloud product, we found three features of heavy rainfall changing associated with aerosols that the rainfall start and peak time occur earlier and the duration becomes longer. The quantitative differences exist between the two indicators, i.e., the statistic differences of above features between clean and polluted conditions are 0.7, 1.0, 0.8 hours based on AOD and 1.4, 3.0, 2.2 hours based on CDNC.

The different relationships of absorbing and scattering aerosols to the diurnal shift were also distinguishable using ultraviolet AI from OMI and reanalysis AOD of two aerosol types (BC and sulfate). The absorbing aerosols (BC) correspond to the earlier start and peak time of heavy rainfall, while the scattering aerosols (sulfate) correspond to the delayed start time and the longer duration. Considering that the moisture has indispensable influence on the rainfall, the role of moisture (SH at 850 hPa) on the heavy rainfall is also investigated, which shows similar with the scattering aerosols (sulfate). Further analysis indicates the duration of heavy rainfall is prolonged in the presence of more sulfate on the relatively dry condition but is shortened on the extremely wet condition.

By comparing the characteristics of cloud macrophysics and microphysics variables, using both AOD and CDNC we found the CF, ice COT, liquid and ice CWP are increased on the polluted condition, but the cloud top height and the ice CER are reduced. Liquid COT and liquid CER are also increased in AOD analysis. Comparing the influences of CDNC which represents CCN and SH at 850 hPa which represents moisture condition respectively on these cloud variables, the cloud properties show similar changes with the increase of CDNC and moisture, but seem more sensitive to the CDNC (CCN), e.g., the liquid & ice COT and CWP increase more in the environment of high CDNC than that of high SH.

According to these results, we speculate that both aerosol radiative effect and cloud effect have impacts on the diurnal variation of heavy rainfall in the BTH region. The heating effect of absorbing aerosols especially BC increases the instability of the lower to middle atmosphere so that generates the heavy rainfall occurrence in advance. And the increased moisture supply and increased aerosols which nucleate more cloud droplets and accumulate more liquid water in clouds, leading to the longer duration of heavy rainfall.

6.2 Discussion

In this study we used two aerosol indicators, AOD and CDNC, which discriminates the pollution levels for different purposes. AOD is a good proxy for the large-scale pollution level, but it stands for the optical feature of aerosols and cannot well represent CCN when we focused on the aerosol-cloud interaction (Shinozuka et al., 2015). CDNC is a better proxy for CCN compared with AOD, which facilitates the study on the cloud changes associated with aerosol pollution. But the retrieved CDNC has larger uncertainties. First, the assumptions in the calculation of CDNC are idealized that CDNC is constant with height in a cloud and cloud liquid water increases monotonically at an adiabatic environment (Grosvenor et al., 2018), but the target of this study is the convective clouds with rainfall that may be not consistent with the adiabatic assumption. Second, as indicated

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by Grosvenor et al. (2018), the uncertainties in the pixel-level retrievals of CDNC from MODIS with 1°x1° spatial resolution can be above 54%, which come from the uncertainties of parameters and the original COT and CER data using in the calculation, and also the influence of heterogeneity effect from thin clouds. To reduce the influence of heterogeneity effect as much as possible, we have attempted to limit the conditions of CF, liquid COT and CER when calculating CDNC in the study. Besides, this study primarily focuses on the relative changes of CDNC, which may be also influenced by the potential systematic biases in the CDNC calculation, but actually reduced the uncertainties of absolute values. Another problem of CDNC in this study is that the CDNC is actually influenced by updraft velocity when we use CDNC to represent CCN since both CCN and updraft velocity could contribute to aerosol activation and increase CDNC (Reutter et al., 2009). We used the vertical velocity at 850 hPa obtained from ERA-interim reanalysis data to investigate the relationship between CDNC and updraft, and the results show that there is no significant correlation found between CDNC and vertical velocity (among all cases or just polluted cases), although the vertical velocity is larger in the polluted cases. We also checked the results of rainfall based on CDNC when limiting the vertical velocity to a certain range (less than 25^{th} percentile, 25^{th} – 75^{th} percentile or more than 75^{th} percentile), and the main conclusion did not change. Besides, the increase of vertical velocity on the polluted days during daytime we think is related to the pollutants (Zhou et al., 2018), which means the pollutants including the aerosols serving as CCN and the updraft are co-varied and they might be the cause and effect for each other. Therefore, we suppose the CDNC could stand for CCN to a certain extent although it might be partially dependent of updraft velocity.

In addition to AOD and CDNC, we also applied ultraviolet AI and AOD of BC/sulfate to identify different types of aerosols. We found that the AI has a weak positive correlation with AOD from MODIS, which indicates the results on absorbing aerosol days might represent the results on polluted days if identified by AOD. To avoid the uncertainty, we re-examined the results using AI when removing the polluted cases identified by AOD, and found the major results are not changed. The comparisons of BC/sulfate AOD cases also have uncertainties because they are retrieved from MACC reanalysis data. Although the above four indicators have their own uncertainties, currently we cannot find more reliable datasets in a long-term observational record. The major findings using these four indices could well identify the changes of rainfall and clouds accompanied with aerosols, but are insufficient to clarify the aerosol effect on clouds and precipitation.

This study has clearly identified the relationship of the aerosol pollution and the diurnal changes of heavy rainfall and associated clouds in the BTH region. However, although this work has attempted to exclude the impacts from the meteorological background particularly circulation and moisture, the observation study still has its limitations on studying aerosol effects on rainfall and clouds: first, the noise and uncertainty of different observational data cannot be avoid, e.g., the misdetection of CF in the satellite product when AOD is large (Brennan et al., 2005; Levy et al., 2013) and the mutual interference between liquid and ice clouds (Holz et al., 2008; Platnick et al., 2017); Second, the meteorological co-variations cannot be completely removed

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thus bring the uncertainties, e.g., the meteorology might have a vital influence on the relationship of AOD and CF (Quaas et al., 2010; Grandey et al., 2013) and the relationship of AOD and CTP (Gryspeerdt et al., 2014a); Third, the different types of aerosols cannot be completely separated, although we used AI index and AOD of BC/sulfate to distinguish the respective effects of absorbing aerosols and scattering aerosols. In addition, we selected the extreme ranges of AOD/CDNC to compare the characteristics of heavy rainfall and associated clouds, which could bring uncertainties that these extreme conditions might be related with totally different microphysical process or meteorological background. So we further checked the results using the middle range of AOD and CDNC such as 25^{th} – 50^{th} percentile versus 50^{th} -75^{th} percentile. The results are basically the same and significant except the change of the peak time of heavy rainfall is not significant based on AOD. The influence of aerosol pollution on the heavy rainfall and clouds may not be linear, but this study using the idea of comparative experiments gives the observational results of the relationships between them. Numerical model simulations are necessarily applied to further study the specific impact of aerosols on the heavy rainfall. And the detailed processes of aerosol effect on the precipitation formation of mix-phased clouds also needs further exploration in our future study.

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Data availability

We are grateful to the National Meteorological Information Centre (NMIC) of the China Meteorological Administration (CMA) for providing hourly precipitation datasets. MODIS aerosol and cloud data were obtained from http://ladsweb.modaps.eosdis.nasa.gov; ultraviolet AI data from OMI was obtained from https://daac.gsfc.nasa.gov/datasets?keywords=OMI&page=1; MACC-II and ERA-interim reanalysis datasets were obtained from http://apps.ecmwf.int/datasets.

Author contributions

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

Competing interests

The authors declare that they have no conflict of interest.

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Tables

Indicator	Source	Begin time	Thresholds	
			25th percentile	75th percentile
AOD	MODIS	2002	0.98	2.00
CDNC (cm^{-3})	MODIS	2002	30.10	91.03
AAI	OMI	2005	0.13	0.52
SAI	OMI	2005	- 0.13	- 0.35
AOD of BC	MACC	2003	0.04	0.06
AOD of sulfate	MACC	2003	0.46	0.87
SH at 850 hPa (g/kg)	ERA-interim	2002	9.96	12.95

Table 1. The indicators of aerosols and moisture used in the study and their sources, begin times and the thresholds (25th and 75th percentiles). The end time of all data is to 2012.

Characteristics of heavy rainfall	Clean		Polluted		Difference		Significance	
	AOD	CDNC	AOD	CDNC	AOD	CDNC	AOD	CDNC
Start time	24.2 (3.9)	24.3 (4.0)	23.5 (4.8)	22.9 (3.9)	- 0.7	- 1.4	P<0.05	P<0.05
Peak time	23.0 (4.0)	22.1 (5.3)	22.0 (4.8)	19.1 (5.7)	- 1.0	- 3.0	P<0.05	P<0.05
Duration	4.0 (2.1)	5.5 (3.3)	4.8 (2.8)	7.7 (4.3)	0.8	2.2	P<0.05	P<0.05
Intensity	164.9 (98.4)	166.0 (89.3)	169.6 (94.3)	162.7 (89.1)	4.7	- 3.3	P>0.1	P>0.1

Table 2. The mean values of start time (units: LST), peak time (units: LST), duration (units: hours) and intensity (units: 0.1mm/hour) of heavy rainfall respectively on the clean and polluted conditions using two indicators of AOD and CDNC, and their differences (polluted minus clean) and significances. The numbers in the brackets stand for the standard deviations on the means. "P<0.05" stands for the difference has passed the significance test of 95%, and "P>0.1" stands for the difference did not pass the significance test of 90%.

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Characteristics of heavy rainfall	AAI	SAI	Difference (AAI-SAI)	Less BC	More BC	Difference (More-Less)	Less sulfate	More sulfate	Difference (More-Less)
Start time	23.4 (4.8)	24.1 (4.4)	-0.7	24.2 (4.8)	23.9 (4.4)	-0.3	24.0 (4.3)	24.5 (4.4)	0.5
Peak time	21.0 (5.3)	22.6 (5.1)	-1.6	23.4 (5.3)	22.3 (4.0)	-1.1	23.2 (4.5)	22.9 (4.8)	-0.3
Duration	5.0 (3.1)	6.0 (3.8)	-1.0	4.8 (2.6)	4.6 (2.7)	-0.2	4.0 (2.1)	5.5 (3.0)	1.5

Table 3. The mean values of start time (units: LST), peak time (units: LST) and duration (units: hours) of heavy rainfall respectively on the conditions with more absorbing aerosols (AAI more than 75th percentile, from OMI), more scattering aerosols (SAI more than 75th percentile, from OMI), less or more BC (AOD of BC less than 25th or more than 75th percentile, from MACC), less or more sulfate (AOD of sulfate less than 25th or more than 75th percentile, from MACC), and their differences. Numbers in the brackets stand for the standard deviations on the means. All differences have passed the significant test of 95%.

Clean/Polluted		CF	CTP	COT		CWP		CER	
				liquid	ice	liquid	ice	liquid	ice
AOD	Clean	62.8 (17.6)	442.3 (149.6)	6.9 (4.5)	6.7 (8.5)	62.8 (36.6)	123.1 (168.9)	16.7 (4.4)	32.0 (8.7)
	Polluted	89.3 (12.9)	487.3 (145.7)	10.0 (5.8)	12.9 (17.0)	96.4 (52.5)	211.3 (279.3)	17.5 (3.5)	29.2 (9.0)
CDNC	Clean	94.5 (6.1)	398.0 (131.7)	8.1 (6.0)	8.7 (10.6)	102.4 (104.3)	171.6 (204.3)	20.4 (2.8)	34.2 (6.0)
	Polluted	97.4 (4.2)	430.8 (135.2)	40.4 (21.5)	33.1 (22.7)	318.2 (213.2)	542.5 (447.8)	12.2 (1.9)	25.4 (8.7)

Table 4. The mean values of CF (units: %), CTP (units: hPa), COT (liquid and ice, units: none), CWP (liquid and ice, units: g/m^2) and CER (liquid and ice, units: μm) from MODIS C6 cloud product on the clean condition (less than 25th percentile) and polluted condition (more than 75th percentile) using two indicators of AOD and CDNC. Numbers in the brackets stand for the standard deviations on the means. Numbers in grey indicate the results of liquid COT & CER are related to the calculation of CDNC. The differences between clean and polluted conditions have all passed the significant test of 95%.

	Group (case number)	CF	CTP	COT		CWP		CER	
				liquid	ice	liquid	ice	liquid	ice
1	Clean, dry (153)	93.8 (6.1)	393.3 (117.3)	7.2 (4.6)	7.6 (9.4)	88.7 (70.6)	149.0 (146.4)	20.4 (3.0)	36.7 (6.6)
2	Polluted, dry (128)	95.6 (5.1)	475.7 (142.8)	50.2 (24.4)	43.4 (19.3)	424.6 (275.5)	793.5 (404.7)	12.6 (2.4)	30.0 (7.0)
3	Clean, wet (155)	*92.7 (7.0)* $p_{1,3}>0.05$	457.4 (191.0)	8.6 (4.7)	10.6 (12.6)	101.9 (64.5)	207.7 (254.1)	*19.8 (2.5)* $p_{1,3}>0.05$	33.2 (4.4)
4	Polluted, wet (194)	97.8 (4.4)	*419.7 (141.0)* $p_{3,4}>0.05$	36.4 (20.6)	28.4 (21.1)	295.9 (208.7)	456.4 (412.1)	*12.5 (2.0)* $p_{2,4}>0.1$	24.4 (7.5)

Table 5. The mean values of CF (units: %), CTP (units: hPa), COT (liquid and ice, units: none), CWP (liquid and ice, units: g/m^2) and CER (liquid and ice, units: μm) in four groups. Numbers in the brackets stand for the standard deviations on the means. Italic numbers in grey represent that the differences are not significant, in which "P>0.05" stands for the difference has passed the significance test of 90% but did not pass the significance test of 95%, and "P>0.1" stands for the difference did not pass the significance test of 90%.

Figures

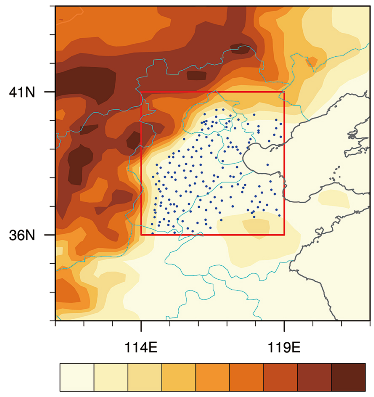

Figure 1. Selected rainfall stations (blue dots) and topography (shading, units: m) in the BTH region (red box, 36–41° N, 114–119° E).

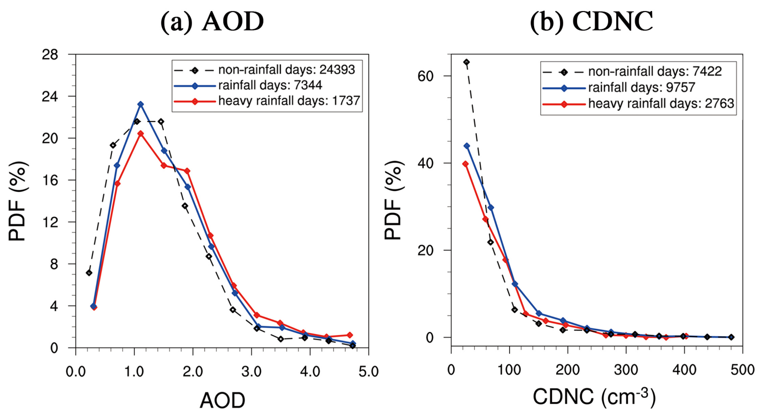

Figure 2. PDF of (a) AOD and (b) CDNC (cm^{-3}) (data from MODIS) on non-rainfall days (black lines), rainfall days (blue lines) and heavy rainfall days (red lines) in southwesterly during early summers from 2002 to 2012. Numbers in the legends denote the sample number.

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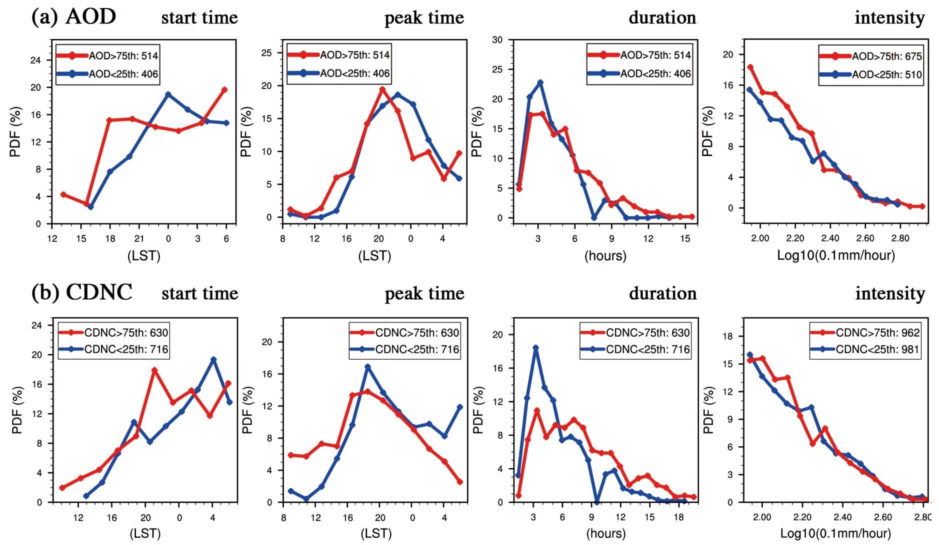

Figure 3. PDF of start time (units: LST), peak time (units: LST), duration (units: hours) and intensity (units: 0.1mm/hour) of heavy rainfall (data from CMA) on selected clean (blue lines) and polluted (red lines) conditions, respectively using indicator of (a) AOD and (b) CDNC (cm^{-3}), during early summers from 2002 to 2012.

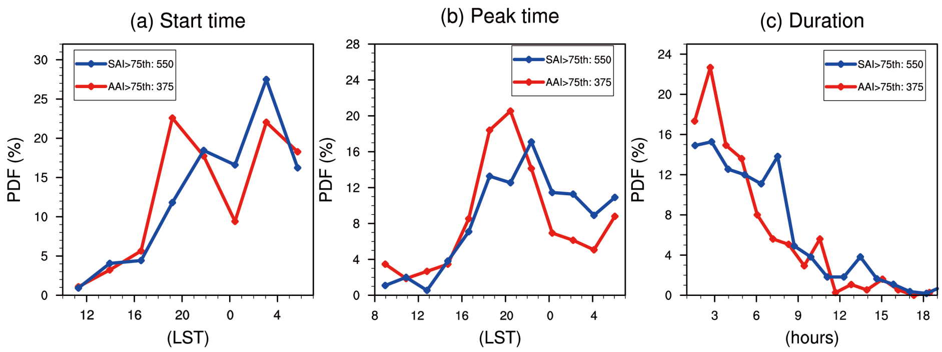

Figure 4. PDF of (a) start time (units: LST), (b) peak time (units: LST), and (c) duration (units: hours) of heavy rainfall on the days with SAI more than 75th percentile (blue lines, data from OMI) and days with AAI more than 75th percentile (red lines, data from OMI), during early summers from 2005 to 2012.

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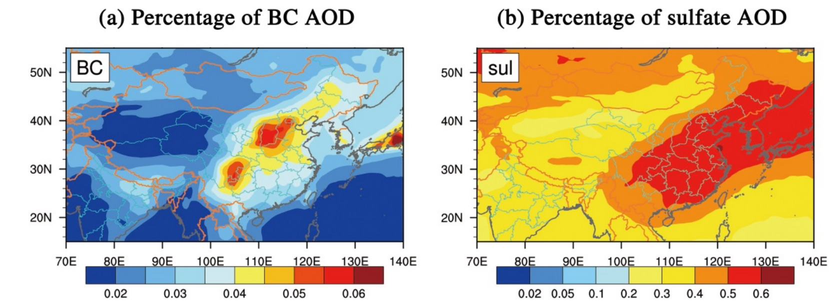

Figure 5. Percentages of AOD for (a) BC and (b) sulfate from MACC reanalysis data in summers (June – August) during 2002 to 2012.

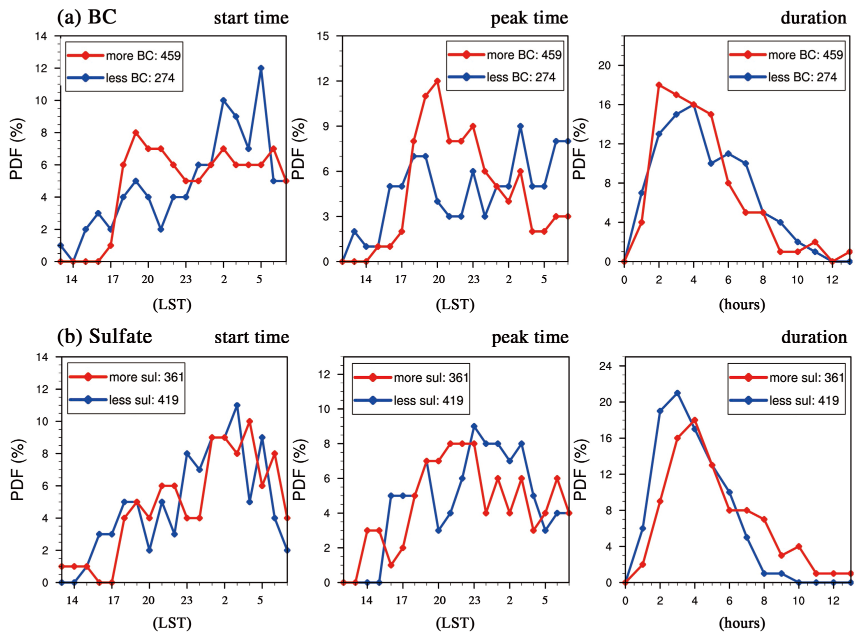

Figure 6. PDF of start time (units: LST), peak time (units: LST) and duration (units: hours) of heavy rainfall on the different conditions of (a) BC and (b) sulfate. Blue/red lines stand for the condition of less/more BC or sulfate (AOD of BC or sulfate less than 25th /more than 75th percentile, data from MACC) during early summers from 2003 to 2012.

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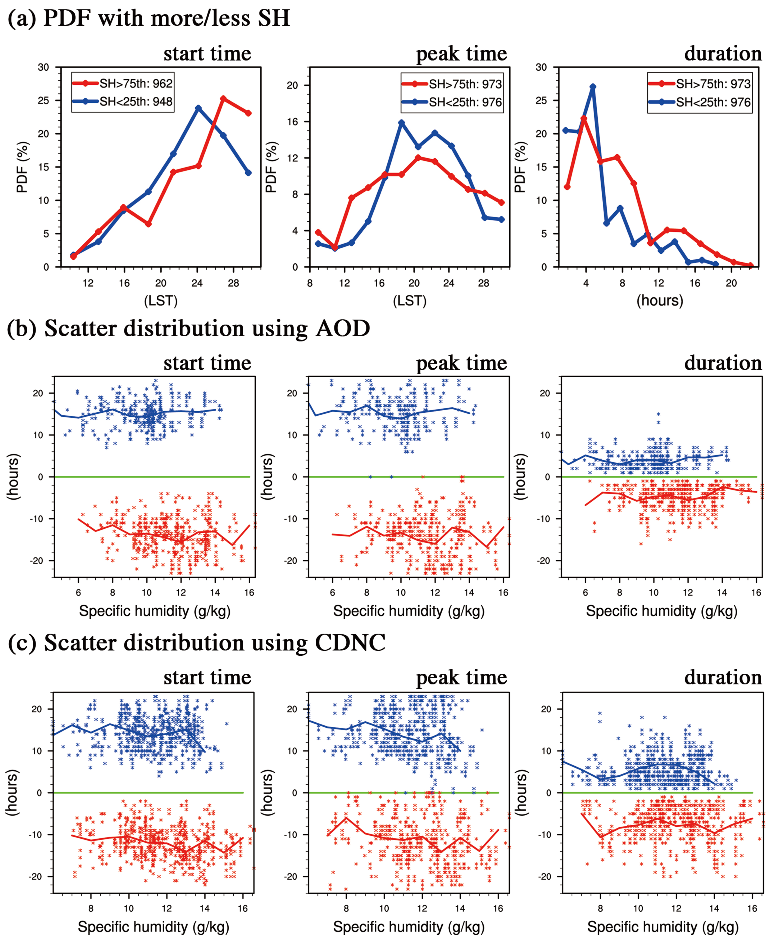

Figure 7. (a) PDF of start time (units: LST), peak time (units: LST), and duration (units: hours) of heavy rainfall with less moisture (blue lines, SH at 850 hPa less than 25th percentile, data form ERA-interim) and more moisture (red lines, SH at 850 hPa more than 75th percentile, data form ERA-interim). (b) and (c) are scatter distributions of SH-start time/peak time/duration for clean cases (blue points) and polluted cases (red points) respectively using AOD and CDNC. Green lines stands for the start/peak time at 8:00 LST or the duration is 0 hours. Positive (negative) values stand for the hours away from 8:00 LST or 0 hours in clean (polluted) cases. Blue (red) lines stand for the mean values of rainfall characteristics at each integer of SH in clean (polluted) cases.

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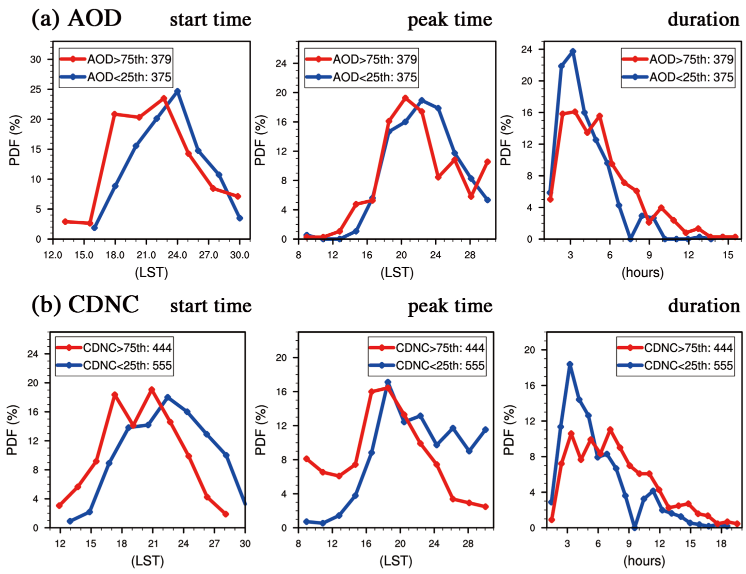

Figure 8. PDF of start time (units: LST), peak time (units: LST), and duration (units: hours) of heavy rainfall on selected clean (blue lines) and polluted (red lines) conditions with SH at 850 hPa (from ERA-interim) less than 75^{th} percentile, respectively using indicator of (a) AOD and (b) CDNC (cm^{-3}), during early summers from 2002 to 2012.

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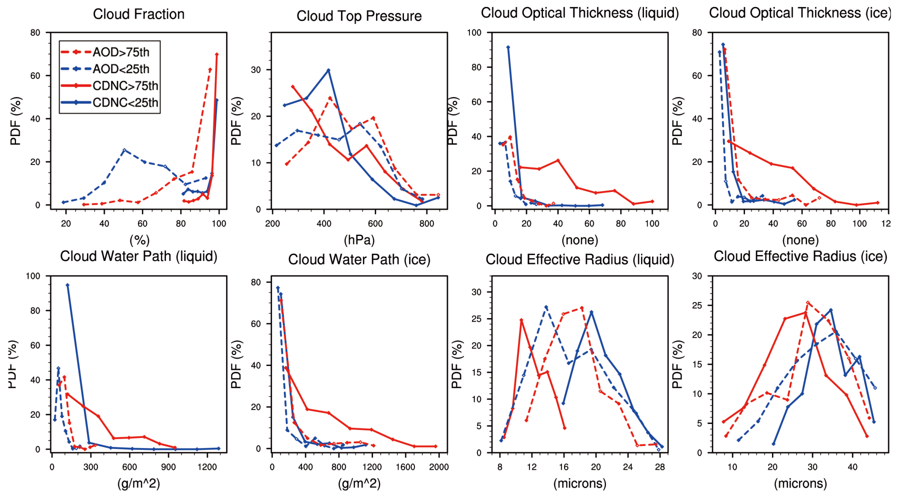

Figure 9. PDF of CF (units: %), CTP (units: hPa), COT (liquid and ice, units: none), CWP (liquid and ice, units: g/m^2) and CER (liquid and ice, units: μm) on selected clean (blue dash lines: AOD<25th percentile; blue solid lines: CDNC<25th percentile) and polluted (red dash lines: AOD>75th percentile; red solid lines: CDNC>75th percentile) heavy rainfall days. All cloud variables are obtained from MODIS C6 cloud product.

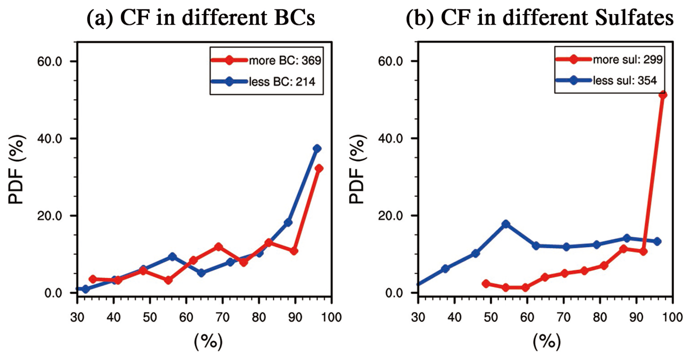

Figure 10. PDF of CF (units: %, data from MODIS) respectively for the conditions of less BC/sulfate (blue lines, AOD of BC/sulfate less than 25th percentile, data from MACC) and more BC/sulfate (red lines, AOD of BC/sulfate more than 75th percentile, data from MACC) cases with heavy rainfall during 10 early summers (2003-2012).

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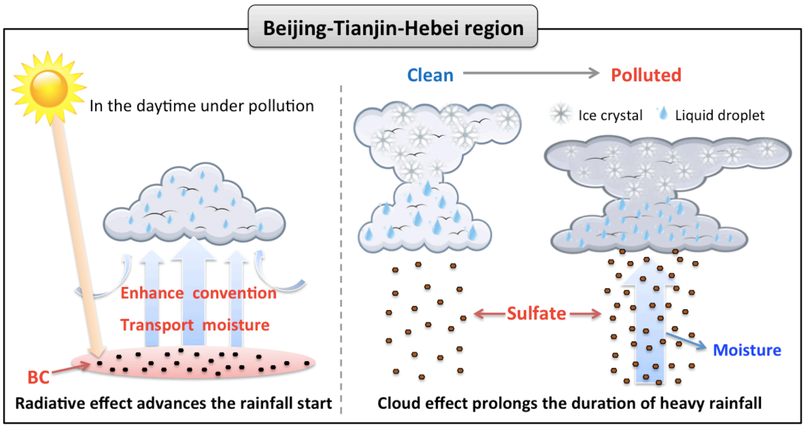

Figure 11. A schematic diagram for aerosol impacts on heavy rainfall over Beijing-Tianjin-Hebei region.

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