Answers to Referee #1's comments

We appreciate very much the constructive comments the reviewer provided, all of which are addressed. The one-to-one correspondence is given below in black.

The authors have done a lot of work redoing the paper, and I applaud their effort.
I have two critical concerns with this paper:
1. Moisture and CDNC/AOD covary in the sample. No multivariate analysis is undertaken and it is possible that covariability between CDNC/AOD and cloud properties or rain statistics are really just moisture variability covarying with precipitation and cloud.
2. The calculation of CDNC here does not acknowledge the numerous sources of retrieval error that may covary with precipitation.

R: Since the effects of aerosols and moisture on precipitation are not linear, multivariate analysis may not provide useful information. Nevertheless, on the concern about the moisture and CDNC/AOD covary, we add Sect. 3.3 to address the effects of moisture on the heavy rainfall.

We agree that the calculation of CDNC needs to be more rigorous and thus we add limiting conditions of CF/COD/CER when calculating CDNC, and add more discussions about the uncertainties of the calculation, see Lines 184-188 in Sect. 2.1.3 and Lines 493-501 in Sect. 5.2. In addition, we also add discussions about the uncertainties of other indicators and associated results in Sect. 5.2.

My central concern- and what may be a critical error in the analysis- is that moisture and pollution covary. The authors reveal this in a later part of the paper. To me this potentially means that all their univariate analysis of rain statistics and cloud statistics as a function of CDNC and AOD are spurious and may just be aliasing onto meteorology. This may not be the case once variability in meteorology is better accounted for, but the authors need to state that moisture and aerosol covary in this region right away and then do all of their analysis in a multivariate way (for example, showing variables as 2D PDFs in the space of moisture and CDNC/AOD/AI instead of 1-D PDFs in the space of just CDNC/AOD/AI).

R: The analysis of rain statistics is not spurious, as fully discussed in our previous study (Zhou et al., 2018) of the sensitivity of rainfall associated with pollution to the moisture and wind speed. In any case, in the revised manuscript we have: added a section to compare the rainfall behaviors associated with moisture and pollution respectively, and, follow the suggestions, illustrated the scatter distribution of moisture-rainfall characteristics relationship of clean and polluted cases (Fig. 6b&c, equivalent to 2D PDF) and discussed their difference, see Sect. 3.3.

The authors have added CDNC, but their approach to this does not incorporate knowledge of the various sources of systematic error in CDNC retrievals from MODIS that we know of (Daniel P. Grosvenor et al., 2018). The authors need to expand their discussion of CDNC, clarify exactly how they are calculating this quantity, and discuss how known covariances between cloud habit, rainfall rate, and CDNC retrieval error might impact their results. This will substantially impact section 5.1 of the MS, but needs to be done as there has been shown to be substantial covariability between cloud morphology and CDNC retrieval error (D. P. Grosvenor & Wood, 2014).

R: We agree and have added discussions on some uncertainties, see Lines 493-501 in Sect. 5.2.

The incorporation of OMI aerosol index is interesting, and supports their work. I do not know anything about these products, but it is less clear how spurious covariability between these retrievals and rainfall could occur than with MODIS AOD/CDNC. I suggest that they incorporate these measurements into their evaluation of rainfall statistics.

R: As shown below, the relationship between aerosol index from OMI and AOD from MODIS show a weak positive correlation. With regard to the co-variability, we have tested the result when removing the cases with AOD more than 2.00, the polluted cases. It shows the consistent result that the heavy rainfall occurs earlier on absorbing aerosol days and the duration is a little longer on scattering aerosol days. We addressed this issue in Sect. 5.2, see Lines 505-508.

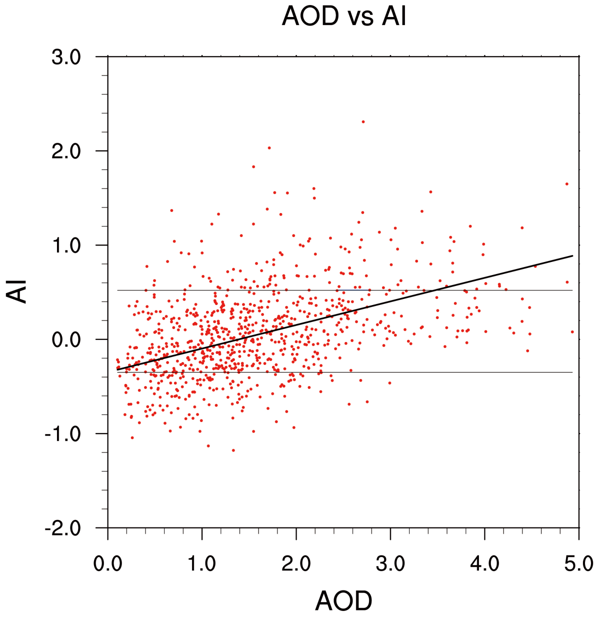

Figure A1. Relationship between AI from OMI and AOD from MODIS on the southwesterly days. The bold line stands for the regression and the thin lines are the thresholds of AI used in the study.

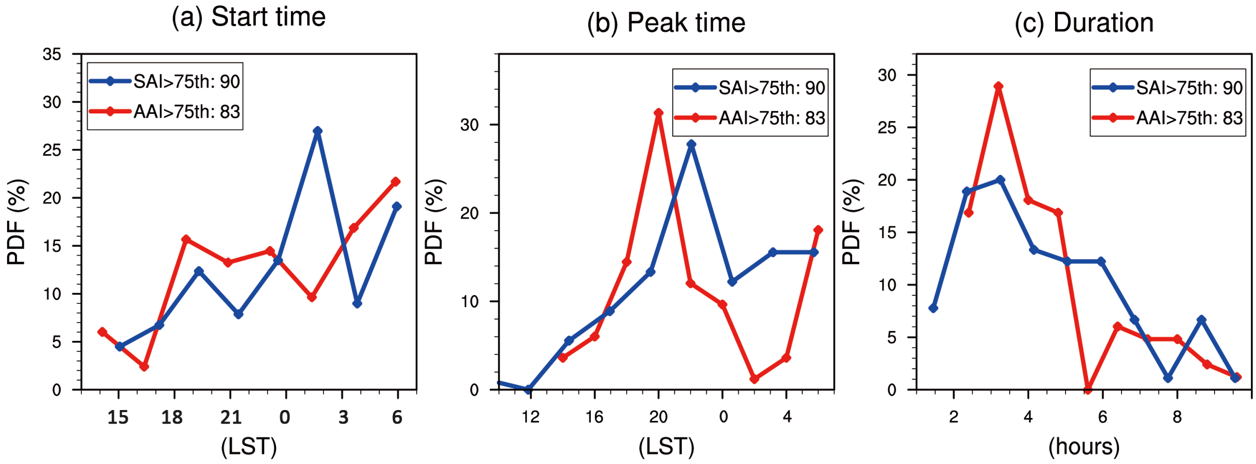

Figure A2. PDF of (a) start time (units: LST), (b) peak time (units: LST), and (c) duration (units: hours) of heavy rainfall on the days that SAI more than 75th tercile (blue lines) and days that AAI more than 75th tercile (red lines) when AOD is less than 2.00, during early summers from 2005 to 2012.

Overall, I think the paper has potential in the use of ground-based data to get around shortcomings in remote-sensing of rain, but some work has to be done on the analysis to acknowledge the information we have about CDNC retrievals and to work up from the assumption that meteorological variability is the leading order driver of cloud and precipitation variability- as opposed to putting it in as an afterthought.

The paper has some difficulty with grammar, but generally the intent of the authors is understandable and I am assuming writing will be tuned up by the ACP proofing staff.

R: We have tried our best to edit the manuscript.

Ln 36: Do the authors mean this paper, or a recent paper they have published?

R: We mean a recent paper we have published (Zhou et al., 2018). The sentence is revised, see Line 36.

Ln 47: Correlation and causation are not the same thing. Changes in cloud statistics covary with the proxies for aerosol used in this study.

Ln 51: It is unclear what this sentence means.

Ln 47: It is unclear what these numbers mean. What was the change in aerosols (AOD/CDNC?)

R: We have re-written the abstract, see Lines 46-51.

Ln 84: What is moisture supply? Is this the RH at some pressure level? I see this isn't described until line 188.

R: We use mean specific humidity at 850hPa (absolute humidity at 850hPa in the last version). This is explicitly mentioned in the abstract and introduction, see Lines 49-50, 105-106.

Ln 134: I would say that it tends to be before heavy rainfall events, however, I find Fig. 2 a little confusing- where did LST 6-13 go? Where there never any rainfall events in that time range? That seems unlikely. If I linearly interpolate between the last point and the first point I can see that there a fair number of rainfall events occurring at 10:30 LST, although not the maximum of the PDF- so I would say 'tends' and not 'always'.

R: To better match other satellite data and meteorological data, we count "a day" to be from 8 LST to 8 LST next day (0 UTC – 24 UTC). The Terra satellite overpass the BTH region is at around 10:30 LST and when it overpasses BTH, the data is almost missing when it is rainy at that time. So it is not that there are never any rainfall events, rather the rainfall data at that time is removed when we use satellite data. In addition to the missing data, it shows that there are less rainfall cases starting in the morning. Shown in the figure below, the only difference between the two lines is that the latter added ' if (.not. is missing(AOD))' in the code. We revised the statement, see Lines 139-142 in Sect. 2.1.2.

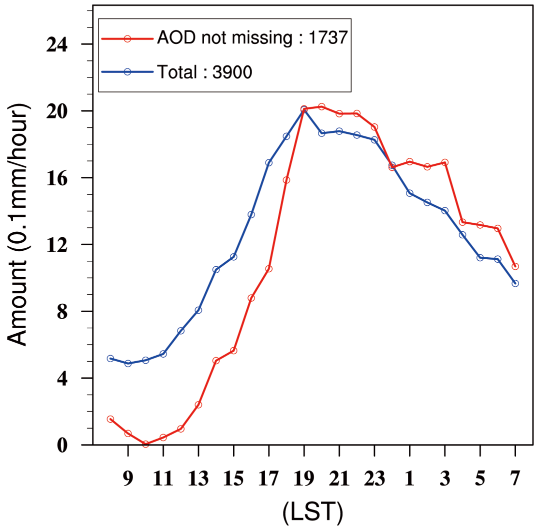

Figure A3. Diurnal variations of rainfall amount (0.1mm/hour) on the total heavy rainfall days (blue line) and heavy rainfall days with AOD record (red line).

Ln 167: What band is used to retrieve reff? It makes a big difference (Daniel P. Grosvenor et al., 2018). Are low (80%) cloud fractions removed? From looking ahead I infer that they are not. This is also important to deal with heterogeneity effects, which are critical since the authors are looking at precipitation (see restrictions applied in (Bennartz, Fan, Rausch, Leung, & Heidinger, 2011)).

R: We have added limiting conditions of CF when calculating CDNC as you suggested, and the results are consistent when removing low (less than 80%) cloud fraction. In addition, we have added discussion about the uncertainty of CDNC, see Lines 184-188 in Sect. 2.1.3 and Lines 493-501 in Sect. 5.2.

Ln 217: As noted in the previous revision, it is unclear if CDNC can be used to stratify into clean and polluted cloud to learn anything about rain. This is because cloud top heterogeneity affects the retrieval of CDNC from MODIS (see Fig 16 of D. P. Grosvenor and Wood (2014)). For example, higher cloud top heterogeneity could go in step with higher rain rates and higher CDNC. The authors need to discuss this source of spurious covariability.

R: We have added limiting conditions of CF/COD/CER when calculating CDNC to reduce the influence of heterogeneity effect, and added discussions about the uncertainties of CDNC in the revised version, see Lines 184-188 in Sect. 2.1.3 and Lines 493-501 in Sect. 5.2.

Ln 238: I think this section is compelling, especially combined with table 2, which shows that high AOD and CDNC tend to lead to the same change in sign for changes in precipitation characteristics. I do recommend that the authors look into the difference in log intensity since the distribution of intensity is clearly non-normal. Can a similar table be made for the AI from OMI and SO4/BC AOD? That would be nice to compare across.

R: We have revised the intensity in Fig. 2 into log intensity but the changes are also non-significant. A table (Tab. 3) made for AI and sulfate/BC AOD is added in the revised manuscript.

Ln 302:

I think that this section looking at cloud changes sorted by CDNC could be removed. The retrievals of CDNC, LWP, CF, CER and COT are all dependent on each other and retrieval errors in one will affect all the others. I am not sure how much this analysis adds to the primary results of the paper, which is (in my reading) the covariability between *precipitation statistics and CDNC/AOD. However, I understand that this analysis helps the authors support their hypothesized mechanism, so I think that if the retrievals in this section are heavily caveated in that none of the variables are truly independent retrievals then that will be acceptable.*

R: This section is revised as follows. Both AOD and CDNC are used to investigate the concurrent cloud changes associated with pollution during heavy rainfall days. Only changes of variables such as CTP, liquid & ice CWP, and ice CER that are independent of CDNC retrieval are compared with the results based on AOD. We have revised this part and added discussions about the uncertainties of cloud change based on AOD and CDNC, see Lines 360-368 in Sect. 4.1 and Lines 521-537 in Sect. 5.2.

Ln 377: In the last part of the section the authors discuss how moisture and CDNC covary in their sample. This means that most of the paper up till now may be looking at spurious covariability. We would expect that meteorology (in this case- moisture) to dominate driving cloud property variability. This also means that the results relating to rainfall are potentially just looking at times when it is more or less moist.

To clarify- if I believe that aerosol has no effect on rain rates, based on what the authors have just told me it seems perfectly reasonable to suspect that meteorology drives everything and that CCN-proxies are just covarying with moisture. Obviously this is an empirical study and they can't fully rule out the possibility that CCN proxies are covarying with some unknown predictor, but the authors need to start out binning by CCN proxy and their meteorological parameter at the beginning instead of introducing it now.

R: In our previous study (Zhou et al., 2018), we have indicated that advance of heavy rainfall still exists when limiting the condition of humidity. Classifying the heavy rainfall by AOD/CDNC and moisture at the beginning will lead to the large difference of case numbers between clean and polluted conditions, which causes more uncertainty to rainfall statistics. Hence, to address the issue you raised, we added a section to compare the moisture effect and aerosol effect on the heavy rainfall, see Sect. 3.3.

Ln 416: Or estimated inversion strength and BC covary.
R: We agree and added this phrase, see Lines 442.

Ln 431: Or estimated inversion strength and SO4 covary- or SO4 is bright and leads to enhanced cloud fraction through misdetection.
R: We modified the statement, see Lines 458-461 in Sect. 5.1. The misdetection of MODIS is possible, although the MODIS C6 product has progress in detect aerosols and clouds (Levy et al., 2013). We added discussions in Sect. 5.2, see Lines 483-486.

Fig 8. Something looks off in this plot and Fig. 6 (top left). The CF doesn't seem to have a very smooth distribution. Why are there so many completely overcast 1x1° days with no transition?
R: Since the daily clouds we investigate are all on the heavy rainfall days, we suppose most cases with more than 90% cloud fraction might be reasonable. The misdetection of CF is also possible, and we added discussion about it, see Lines 484-485 in Sect. 5.2.

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Answers to Referee #2's comments

Thanks for the helpful comments. The point-by-point response is given below in black.

I thank the authors for the work they have put into revising the paper, it is much improved over the previous version, although there are a number of things that I am still concerned about.

The linking of aerosol properties to the diurnal variation of aerosol remains a nice result to base this study around, but the correlation between cloud and aerosol properties shown in later parts of the work is subject to a number of meteorological covariations. Just controlling for the humidity has been shown to be insufficient to isolate an aerosol effect (Quaas and Boucher, 2012). With this in mind, Section 5 is useful in enumerating the cloud properties that are correlated to CDNC, but I am not sure it demonstrates a causal role of aerosol. I am not sure that it is vital to explain the rainfall results from the first part of the paper. Perhaps this section could be presented relationships between cloud and aerosol properties, rather than evidence of an aerosol impact.

R: We agree, and the subtitles and some statements in Sect. 5 (now Sect. 4 in the revised manuscript) have been revised.

Main points:
I like the use of the CDNC as an aerosol proxy, but I think that the authors need to do more to demonstrate that it is applicable in this case. It is not just a drop-in replacement for AOD and has a number of specific biases.

The CDNC calculation is only applicable in adiabatic clouds. In general, convective clouds are not adiabatic and a precipitating cloud is unlikely to be. This may not be a large factor in this study, but it should be considered and factored into the discussion/conclusions. There are a number of other potential biases that may affect this work. Grosvenor et al. (2018) is a good summary.

Additionally, is the CDNC calculation done using the 1 by 1 degree mean data? As a non-linear calculation, this can strongly impact the ``retrieved'' CDNC. The MODIS L3 product includes a cloud optical depth-effective radius joint histogram which can be used to better calculate the CDNC (e.g. Quaas et al., 2008; Grandey and Stier, 2010)

R: The references are useful. We agree that large uncertainties exist for the derived CDNC (Daniel P. Grosvenor et al., 2018). The CDNC in the study is calculated using 1° x 1° data from MODIS L3 product. As indicated by Grosvenor et al. (2018), the uncertainties in CDNC from MODIS with 1° x 1° spatial resolution can be 54% or even more. However, we assume the relative differences of CDNC at different time and location should be with much less uncertainties, and they are the main information we adopted. To make the calculation of CDNC more rigorous, we have added limiting conditions of CF/COD/CER when calculating CDNC to reduce the uncertainties, and we added some discussions about the uncertainties of the calculation, see Lines 185-188 in Sect. 2.1.3 and Lines 493-501 in Sect. 5.2.

As the CDNC is calculated from the CER and COD, it is not clear to me that the CDNC-CER and CDNC-COD relationships are useful. I have similar concerns about the CDNC-LWP relationship, which is difficult to interpret even in low-level liquid clouds (e.g. Sato et al, 2018; Gryspeerdt et al., 2018b). In this case, I think that Fig. 7 is just reproducing the assumptions used to calculate the CDNC and LWP (if the CER and CDNC are known, then the LWP is uniquely identified - at least at a pixel level).

R: The cloud section is revised to use both AOD and CDNC indicators to investigate the concurrent cloud

changes during heavy rainfall days. Only changes of variables such as CTP, liquid & ice CWP, and ice CER that are independent of CDNC retrieval are compared with the results based on AOD. We have revised this part and added discussions about the uncertainties of cloud change based on AOD and CDNC, see Lines 360-362 in Sect. 4.1 and Lines 532-537 in Sect. 5.2. And Fig.7 is deleted.

Finally, is it clear what biases in the CDNC might be caused by the addition of thin overlying ice cloud? The authors are considering very complex situations where this may be an issue in a way that it is not for studies of liquid clouds.

R: Since the liquid CER and liquid COD from MODIS which calculate the CDNC might be affected by overlying ice cloud, the CDNC might be affected as well. We added discussion about it in the revised version, see Lines 483-486 in Sect. 5.2.

Minor points:
L188 - What are the advantages of the AH here compared to a more common meteorological value such as specific humidity (easily available from ERA-Interim)?
R: We have changed the AH into SH in the revised manuscript, their results show consistent features.

L218 - Absorbing aerosol index is dependent on the altitude of the aerosol layer. What is assumed in this work?
R: We have compared the difference of aerosol profiles obtained from MACC reanalysis data between selected absorbing aerosol days and scattering aerosol days, although the profiles from MACC may be not authentic. The profiles of different aerosols (BC, OC, sulfate and dust) show similar characteristics that the maximums occur at the similar altitude (around 900-1000 hPa). Hence, we assume that the altitude of aerosol layer has almost no influence on the results analyzed by aerosol index in the study.

L406 - How does decreasing the supersaturation reduce the strength of the freezing process? The supersaturation over ice is higher than over liquid. There are some studies which have noted an aerosol relationship to observations of mixed phase and ice cloud that might help explain this result (Chylek et al., 2006; Zhao et al., 2018; Gryspeerdt et al., 2018)
R: Thanks for the references. We have revised the explaination, see Lines 526-528 in Sect. 5.2.

L446 - There have been many studies looking at the impact of BC on precipitation, perhaps they might be helpful in interpreting these results (e.g. Fan et al., 2008; O'Gorman et al., 2011). The role of BC is expected to change with altitude. Is it clear that the BC here is all in the boundary layer?
R: Thanks for the references. Since reliable observational aerosol vertical data are not available, we use MACC and MERRA2 reanalysis data (Fig. 3e in Zhou et al., 2018) to provide this information: the height of BC is around 700-1000 hPa (around 0-3000m from surface). The boundary layer height on the polluted days (based on AOD) is below 1600 m in average during daytime thus there might be BC above the boundary layer. Our results show that under pollution, the upward motion is enhanced, the boundary layer is more stable but the lower to middle atmosphere (850-500 hPa) is more unstable, which may be related to the heating effect of BC (Zhou et al., 2018).

L462 - This might be due to a change in LWP/cloud depth with changing AOD. However, it is not clear if that change in cloud depth could be attributed to aerosols.
R: We agree and revised the statements, see Lines 534-537 in Sect. 5.2.

Figures - The text on many of the plots is very small.
R: We have modified the plots.

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

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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 the effects of aerosols on heavy rainfall, and the concurrent changes of cloud properties. For heavy rainfall, three distinguished characteristics are identified: *earlier start time*, *earlier peak time*, and *longer duration*. The quantitative values of these changes are however sensitive to the choice of pollution indicators: 0.7, 1.0 and 0.8 hours based on aerosol optical depth (AOD); and 2.1, 4.2 and 2.4 hours based on cloud droplet number concentration (CDNC). In-depth analysis suggests that the characteristics of earlier in both start time and peak time occur in the presence of black carbons (absorbing aerosols) while the longer duration is attributed to sulfates (scattering aerosols). Because of its close relevance to changes in heavy rainfall, we also examined changes of clouds. Significant increases in cloud fraction, cloud top pressure, the liquid/ice cloud optical thickness and cloud water path are found. However, changes in cloud microphysics show different responses between AOD and CDNC analyses. While decreases in ice cloud effective radius are found in both analyses, the liquid cloud effective radius is increased in AOD analysis but decreased in CDNC analysis. The effect of moisture (specific humidity at 850 hPa) on heavy rainfall and clouds was also studied and more moisture tends to increase rainfall duration. Finally, the mechanisms which may explain the aerosol effects are discussed and hypothesized.

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

1. Introduction

Aerosols modify the global hydrologic cycle through both radiative effect (direct effect) and cloud effect (indirect effect) (IPCC, 2013). On the one hand, through absorbing or scattering solar radiation, aerosols can lead to the air aloft heating (e.g. Jacobson 2001; Lau et al. 2006) or the surface cooling (Lelieveld and Heintzenberg 1992; Guo et al. 2013; Yang et al., 2018), which changes the atmospheric vertical static stability and modulates rainfall (e.g. Rosenfeld et al. 2008). On the other hand, water-soluble aerosols serving as cloud condensation nuclei (CCN) could affect the warm-rain processes 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). Beijing-Tianjin-Hebei (BTH) region is the heaviest aerosol polluted area in China and concerns have been raised about the aerosol-radiation-cloud-precipitation interaction over this region. The impact of aerosols on light rainfall or warm-rain processes over BTH region almost reaches consistent agreement (e.g., Qian et al., 2009), but aerosol effects on the heavy convective rainfall in this region still have large uncertainties (Wang et al., 2009; Guo et al., 2014; Wang et al., 2016).

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

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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., 2014), 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 (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 concurrent 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., 2014; 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)? (2) how do aerosols influence the concurrent 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 the concurrent 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

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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, including the distinct characteristics of rainfall diurnal variation on clean/polluted conditions; the different behaviors of heavy rainfall diurnal variation along with the change of two 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 pollution and examines the influences of CCN and moisture on the cloud properties. Section 5 makes a discussion on the distinct roles of aerosol radiative effect/cloud effect on the behaviors of heavy rainfall diurnal variation, as well as the uncertainties of different indicators and associated distinct results. Conclusion will be given in Sect. 6.

2. Data and methodology

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 data

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.

2.1.2 Aerosol data

AOD is a proxy for the optical amount of aerosol particles in a column of the atmosphere and serves as one of indicators 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%),

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(+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. As shown in Fig.2, 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.

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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). 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. MACC-II data covers the period of 2003 to 2012.

2.1.3 Cloud data

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.

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CDNC is retrieved as the proxy for CCN and also another 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:

$$\mathrm{CDNC} = \frac{C_w{}^{1/2}}{k}\frac{10^{1/2}}{4\pi\rho_w{}^{1/2}}\frac{\tau^{1/2}}{R_e{}^{5/2}} \qquad (1)$$

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 onboard the Terra satellite. 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 COD 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

Other meteorological factors, including wind, temperature, pressure and SH, were obtained from the ERA-Interim reanalysis datasets with 1°x1°horizontal resolution and 37 vertical levels at six-hour intervals. 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

2.2.1 Method of interpolation

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

2.2.2 Selection of sub-season and circulation

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Consistent with our previous work, we focused on early summer (1 June to 20 July) before the large-scale rainy season starts, 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).

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

With the circulation of southwesterly, 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). We used two indicators to distinguish the clean and polluted conditions, which are AOD and CDNC. The 25th and 75th AOD/CDNC of the whole rainfall days are used as the thresholds of clean and pollution condition, and the values are shown in Tab.1. It shows that there are 514 cases of heavy rainfall on polluted days and 406 cases of that on clean days when using AOD, and 630/716 cases on polluted/clean condition when using CDNC.

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 as shown in Tab.1. We used AAI/SAI more than 75th as the extreme circumstances of absorbing aerosols and scattering aerosols to compare their impacts on heavy rainfall. The case 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 of itself in all rainy days, and cases with less BC/sulfate when that is less than the 25th of itself in the same condition. Accordingly, we selected 459 cases with more BC and 274 cases with less BC with heavy rainfall. Similarly, 361 cases with more sulfate and 419 cases with less sulfate with heavy rainfall were selected.

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 rainy day is larger than 75th tercile of the whole rainy days, and chose dry cases when SH on that day is less than the 25th tercile of the whole rainy days (the thresholds are shown in Tab. 1).

2.2.4 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 between the different groups of aerosol conditions.

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3. Relationship between aerosol pollution and diurnal variation of heavy rainfall over BTH

3.1 Distinct characteristics of heavy rainfall diurnal variation associated with aerosol pollution

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. 2a). 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. 2a, 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 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 6 hours earlier on the polluted days, shifting from around 0:00 LST on the clean days to the 18:00 LST (Fig. 2a). 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. 2a, 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. 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) to examine the above-mentioned results. As a result, the similar changes of heavy rainfall can be well exhibited in CDNC analysis as shown in Fig. 2b. 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. Since the difference of rainfall intensity is not significant in this study, the following analysis only focuses on studying the start time, peak time and duration of heavy rainfall along with aerosol pollution.

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3.2 Distinct behaviors of heavy rainfall diurnal variation associated with two different types of aerosols

Using the indicator of AI, we further investigated 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. 3. 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 (Fig. 3a), with 0.7 hours advance in average (Tab. 3). Similarly, the rainfall peak time also shows earlier on absorbing aerosol days (Fig. 3b), 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. 4a) 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. 4b) 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. 2.2.3. The PDF of the start time, peak time and duration of heavy rainfall in the cases with higher and lower amount of BC are shown in Fig. 5a, respectively. The most striking result is that the maximum frequency of rainfall start time in high BC cases evidently shifts earlier (Fig. 5a). Meanwhile, the mean peak time in high BC cases shows 1.1 hour earlier than that in low BC cases (Tab. 3). And the duration of heavy rainfall is slightly shortened by the averaged 0.2 hours in high BC cases. The features in high BC cases are consistent with the above absorbing aerosol effect. 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 high sulfate cases also exhibit similar with the above scattering aerosol effect, except for the peak time that shows later in scattering aerosols but a little earlier in high sulfate cases (Tab. 3).

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3.3 Behavior comparisons of heavy rainfall diurnal variation influenced by moisture and aerosol.

Moisture supply is an indispensable factor for the precipitation formation. 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 got the rainfall characteristics in more humid (more than 75th tercile) and less humid (less than 25th tercile) environments on the heavy rainfall days regardless of the aerosol condition, as shown in Fig. 6a. The results show the start time of heavy rainfall is delayed by 0.9 hours, the peak time is 0.6 hour earlier and the duration is prolonged by 2.0 hours in average in the more humid environment, which is similar with the result on the condition of more sulfate. Besides, the same results are obtained with different moisture indicator, e.g. the 850 hPa absolute humidity. These results indicate the advance of heavy rainfall start time on polluted days is not caused by more moisture supply, while the longer duration and earlier peak in high sulfate cases might be related to the increased moisture supply.

We also investigate the characteristics of low-level moisture and rainfall behavior distribution in clean and polluted cases respectively using AOD and CDNC (Fig. 6 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 polluted cases, indicating that the polluted cases have 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 polluted cases. For example, when the amount of 850 hPa SH is between 8-12 g/kg, the start &peak time in polluted cases show significant earlier and the duration exhibits slightly increased compared with that in clean cases.

The above results indicate that the advance of heavy rainfall start and peak time in polluted cases might be weakly related to the moisture effect, but the moisture could obviously prolong the duration of heavy rainfall (Fig. 6a). Because the diurnal change of heavy rainfall with more moisture is similar with the behaviors of heavy rainfall with scattering aerosols especially sulfate, we cannot figure out their individual role in this section.

4 Relationship between aerosol pollution and concurrent changes of cloud properties associated with heavy rainfall diurnal variation

4.1 Concurrent changes of cloud properties along with heavy rainfall diurnal variation on clean and

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polluted conditions

To understand the cloud effect of aerosols during heavy rainfall diurnal variation, we need to recognize the concurrent cloud characteristics on 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, as shown in Fig. 7.

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Using AOD as the pollution indicator, 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, which increases 42.2% (Tab. 4). The average CTP on the polluted condition is 487.3 hPa, which is larger than 442.3 hPa on the clean condition and increases 10.2%, 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. 7. Both liquid and ice COT on polluted condition exhibit significant increases compared with that on clean condition. The mean amount of liquid COT is increased by 44.9% and ice COT increases by 92.5% (Tab. 4). Similar with COT, the amount of liquid and ice CWP also increase on polluted condition, which increase by 53.5% and 71.6% respectively. In addition, the liquid CER is increased by 4.8% and the ice CER is decreased by 8.8% on the polluted days. The differences of above cloud properties between clean and polluted cases have all passed the 95% statistical confidence level.

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Using CDNC as another pollution indicator, the above-mentioned changes of cloud properties are consistent with that using AOD, except for liquid CER (Fig. 7). The PDF of liquid CER on the polluted condition shifts to larger size based on AOD but smaller size based on CDNC. The inconsistency of liquid CER using two indictors might be due to the calculation method of CDNC which is not independent on the CF, liquid COD and liquid CER. 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 8.2%, 280.5%, 210.7% and 216.1% 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 25.7% on the polluted condition based on CDNC. We noticed that the changes of the 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.

According to the above comparison, the concurrent changes of cloud properties along with heavy rainfall diurnal variation show consistent results using the two pollution indicators (AOD and CDNC). The pollution corresponds to the increase of CF, COT and CWP both for liquid and ice, but the decrease of cloud top height (the increase of CTP corresponds to the decrease of cloud top height) and ice CER. The changes of liquid CER are opposite between the two indicators, but it might be due to the potential negative correlation between liquid CER and CDNC in the CDNC retrieval. Since we cannot distinguish the liquid part of mix-phased

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clouds from liquid (warm) clouds in the observation, the changes of liquid cloud properties above 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.

4.2 Influences of CCN and moisture on the cloud properties

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 aerosol. 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 aerosols on clouds and its sensitivity to moisture, we purposely investigated the changes of above cloud properties with 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 rainfall day less/more than 25th/75th tercile of the CDNC among the heavy rainfall days, and similarly, the "dry/wet" refers to the SH on that rainfall day less/more than 25th/75th tercile of itself among the heavy rainfall days. The average CDNC is 2168.7 cm^{-3} on the dry condition and 2168.1 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 remain 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 cloud properties. The changes of these cloud variables are the same as that in Sect. 5.1, that the CF, COT and CWP both for liquid and ice are increased on the polluted condition, while the cloud top height and liquid & ice CER are decreased based on CDNC. Among these variables, the liquid & ice COT and CWP are especially larger on the polluted condition, which are 5-6 times larger than that on the clean condition. The liquid CER on polluted condition also changes evidently, which becomes almost a half of that on clean condition. On the wet condition, comparing the group 3 and 4, the changes are similar that the CF, COT and CWP both for liquid and ice are increased and the liquid and 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 the CDNC on the dry condition. The above comparisons indicate that with the increase of CDNC (which stands for CCN), the CF, COT and CWP are increased while the CER is decreased for both liquid and ice clouds regardless of the moisture amount.

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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 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 does not overcome the influence of CCN. The change of liquid CER is not significant on the same clean condition, but the ice CER is significantly decreased with the increase of moisture. 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 on COT/CWP.

The results above indicate that both CCN and moisture have impacts on cloud properties. They both contribute to the increase of CF, COT and CWP, in which the influence of CCN on COT and CWP are significantly larger than moisture. The increase of either CCN or moisture corresponds to the increase of CTP. But when the CCN and moisture increase simultaneously, the CTP becomes smaller. Both CCN and moisture correspond to the decrease of ice CER, while only CCN corresponds to the significant 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 above results, and found most cloud variables show the similar changes with the 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 or moisture but more sensitive to the former, the results in Sect. 5.1 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.

5. Discussion

5.1 Different roles of aerosol radiative effect and cloud effect in heavy rainfall

In Sect. 3 we found that the heavy rainfall has earlier start time 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. 3a&5a). We also compared the effect of BC on the associated clouds. Figure 8a shows the CF larger than 90% rarely occurs in high BC environment, which might be associated with the semi-direct effect of BC (IPCC, 2013) or estimated inversion strength and BC covary. This result indicates the influence of BC on the heavy rainfall in Fig. 5a is mainly due to the radiative effect rather than

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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.

The delayed start of heavy rainfall with scattering aerosols in Fig. 3a and higher sulfate in Fig. 5b 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 8b does shows that in contrast with BC, the CF larger than 90% is significantly increased in the high 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 both the scattering aerosol cases and high sulfate cases (Fig 3c&5b). Since the heavy rainfall shows the similar changes with the increase of sulfate and moisture, we currently cannot separate their respective roles in this study. 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 effect 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. 9) 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 upward motion and accelerates the formation of cloud 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 moisture effect. To further distinguish the individual effect, we need to conduct numerical model simulations in our future study.

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5.2 Uncertainties of different indicators and associated distinct results

The gauge-based hourly precipitation data used in this study is more reliable than other observational and reanalysis precipitation data. In contrast with precipitation datasets, the observation of aerosols and clouds from MODIS might have larger uncertainties, e.g., which come from the misdetection of CF when AOD is large (Brennan et al., 2005; Levy et al., 2013) or the mutual interference between liquid and ice clouds (Holz et al., 2008; Platnick et al., 2017).

In this study, we used two pollution 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 studied the aerosol-cloud interaction (Shinozuka et al., 2015). The value of AOD is also influenced by moisture condition, which is aerosol humidification effect (Twohy et al., 2009; Altaratz et al., 2013). Therefore, we comprehensively analyzed the moisture effect on the results. CDNC is a better proxy for CCN, 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). Second, as indicated 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 COD 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 COD and CER when calculating CDNC in the study. Besides, this study primarily applied the relative changes of CDNC, which actually reduced the uncertainties of absolute values.

We applied ultraviolet AI and AOD of BC/sulfate to identify different types of aerosols. The AI datasets from OMI, which can distinguish the absorbing aerosols and scattering aerosols, also have uncertainties especially for the near-zero values. Hence, we only compare the extreme circumstances of absorbing aerosols and scattering aerosols. We also found 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, and the major findings can be well shown in these four indices.

Using AOD and CDNC we have drawn the same conclusion that the heavy rainfall occurs in advance and the duration is prolonged under pollution (Fig. 2). We found the AOD and CDNC only have a weak positive

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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 CDNC cases exhibit more evident changes of rainfall behaviors in average than that using AOD. The difference of cases selected by the two indicators might due to the non-linear relationship of CCN and pollution that the CCN won't continuously increase when aerosol loading is huge (e.g., Jiang et al., 2016), or due to the misdetection of AOD and the calculation uncertainty of CDNC. Since both the two indicators have their uncertainties, we cannot say the result of which one is more reliable.

Most cloud properties also exhibit the consistent changes using CDNC and AOD. First, the CF, COT and CWP both for liquid and ice clouds 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. We speculate that the earlier start of the precipitation process could inhibit the vertical growth of clouds shown as in Fig. 2. Third, the ice CER decreases under pollution using both AOD and CDNC, which could be ascribed to that 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). Currently the detailed physical processes of cold clouds and mixed-phase clouds are not clear, 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.

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However, the different result occurs for liquid CER between using AOD and using CDNC. The liquid CER is decreased when CDNC increases but increased when AOD increases (Fig. 7). The former is actually the natural result of the negative relationship between CDNC and liquid CER in the calculation. And the latter 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. Therefore, the changes of liquid CER with pollution have some uncertainties.

7. 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 two indicators that are AOD from MODIS aerosol product and retrieved CDNC from MODIS cloud product, we found three features of rainfall changing by 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 roles of absorbing aerosols and scattering aerosols in modifying the

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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 time and peak time of heavy rainfall, while the scattering aerosols (sulfate) correspond to the delayed start time and the longer duration. To distinguish the influence of aerosols, the influence of moisture (SH at 850 hPa) on the heavy rainfall is also investigated, which shows similar with the scattering aerosols (sulfate). By comparing the characteristics of cloud macrophysics and microphysics variables, using both AOD and CDNC we found the CF, COT (liquid and ice), CWP (liquid and ice) are increased on the polluted condition, but the cloud top height and the ice CER are reduced. Comparing the influence of CCN and moisture respectively on these cloud variables, the cloud properties show similar changes with the increase of CCN and moisture, but seem more sensitive to the CCN.

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, lead to the longer duration of heavy rainfall.

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

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.

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Competing interests

The authors declare that they have no conflict of interest.

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Tables

Indicator	Data from	Time	Clean/less (< 25th)	Polluted/more (>75th)
AOD	MODIS	2002-2012	0.98	2.00
CDNC (cm^{-3})	MODIS	2002-2012	952.06	2878.58
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
SH at 850 hPa (g/kg)	ERA-interim	2002-2012	9.96	12.95

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

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**	**24.3**	**23.5**	**22.9**	- 0.7	- 1.4	**P<0.05**	**P<0.05**
Peak time (LST)	**23.0**	**22.1**	**22.0**	**19.1**	- 1.0	- 3.0	**P<0.05**	**P<0.05**
Duration (hours)	**4.0**	**5.5**	**4.8**	**7.7**	+ 0.8	+ 2.2	**P<0.05**	**P<0.05**
Intensity (0.1mm/hour)	**164.9**	**166.0**	**169.6**	**162.7**	**+ 4.7**	**- 3.3**	**P>0.1**	**P>0.1**

Table 2. The average 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 the differences and significances between clean and polluted conditions. "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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Indicator	Da
AOD	M
CDNC (cm^{-3})	M
AAI	
SAI	
BC AOD	M
Sulfate AOD	M
Absolute humidity at 850 hPa (g/m^3)	ERA

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Characteristics of heavy rainfall	Average of c condition	
	AOD	C
Start time (LST)	**24.2**	
Peak time (LST)	**23.0**	
Duration (hours)	**4.0**	
Intensity (0.1mm/hour)	**164.9**	**1**

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Characteristics of heavy rainfall	AAI	SAI	Difference (A-S)	Less BC	More BC	Difference (M-L)	Less sulfate	More sulfate	Difference (M-L)
Start time (LST)	**23.4**	**24.1**	**-0.7** $P<0.05$	**24.2**	**23.9**	**-0.3** $P<0.05$	**24.0**	**24.5**	**0.5** $P<0.05$
Peak time (LST)	**21.0**	**22.6**	**-1.6** $P<0.05$	**23.4**	**22.3**	**-1.1** $P<0.05$	**23.2**	**22.9**	**-0.3** $P<0.05$
Duration (hours)	**5.0**	**6.0**	**-1.0** $P<0.05$	**4.8**	**4.6**	**-0.2** $P<0.05$	**4.0**	**5.5**	**1.5** $P<0.05$

Table 3. The average values of start time (units: LST), peak time (units: LST) and duration (units: hours) of heavy rainfall respectively on the conditions with absorbing aerosols, scattering aerosols, less BC, more BC, less sulfate and more sulfate, and their differences and significances.

Clean/Polluted	CF	CTP	COT (liquid)	COT (ice)	CWP (liquid)	CWP (ice)	CER (liquid)	CER (ice)
Clean (AOD)	**62.8**	**442.3**	**6.9**	**6.7**	**62.8**	**123.1**	**16.7**	**32.0**
Polluted (AOD)	**89.3**	**487.3**	**10.0**	**12.9**	**96.4**	**211.3**	**17.5**	**29.2**
Difference [P – C] **Percentage [(P-C)/C]**	26.5 42.2%	45.0 10.2%	3.1 44.9%	6.2 92.5%	33.6 53.5%	88.2 71.6%	0.8 4.8%	-2.8 -8.8%
Clean (CDNC)	**94.5**	**398.0**	**8.1**	**8.7**	**102.4**	**171.6**	**20.4**	**34.2**
Polluted (CDNC)	**97.4**	**430.8**	**40.4**	**33.1**	**318.2**	**542.5**	**12.2**	**25.4**
Difference [P – C] **Percentage [(P-C)/C]**	2.9 3.1%	32.8 8.2%	32.3 398.8%	24.4 280.5%	216.0 210.7%	371.0 216.1%	-8.2 -40.2%	-8.8 -25.7%

Table 4. The average 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) on clean condition and polluted conditions using two indicators of AOD and CDNC, and their differences (polluted minus clean) and percentages. The differences have all passed the significant test of 95%.

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Group (case number)	CF
1.Clean, dry (140)	**67.9**
2.Polluted, dry (75)	**73.1** $0.05<p_{1,2}<0.1$
3.Clean, wet (191)	**85.8**
4.Polluted, wet (338)	**97.5**

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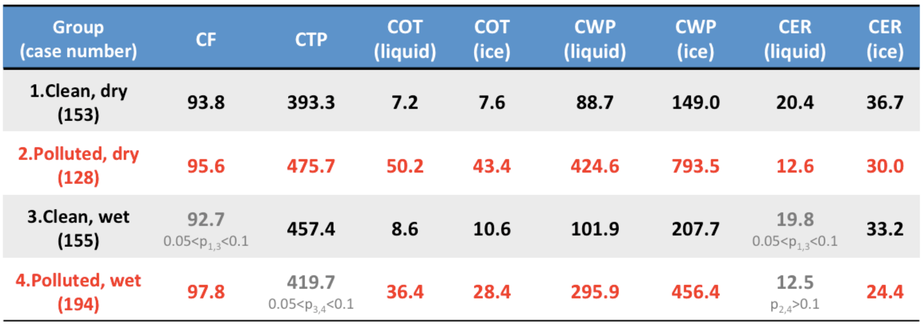

Group (case number)	CF	CTP	COT (liquid)	COT (ice)	CWP (liquid)	CWP (ice)	CER (liquid)	CER (ice)
1.Clean, dry (153)	93.8	393.3	7.2	7.6	88.7	149.0	20.4	36.7
2.Polluted, dry (128)	95.6	475.7	50.2	43.4	424.6	793.5	12.6	30.0
3.Clean, wet (155)	92.7 $0.05<p_{1,3}<0.1$	457.4	8.6	10.6	101.9	207.7	19.8 $0.05<p_{1,3}<0.1$	33.2
4.Polluted, wet (194)	97.8	419.7 $0.05<p_{3,4}<0.1$	36.4	28.4	295.9	456.4	12.5 $p_{2,4}>0.1$	24.4

Table 5. The average 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. Grey numbers represent that the differences are not significant, in which “0.05<P<0.1” 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%.

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Figures

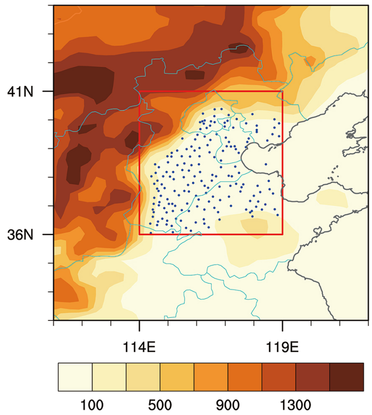
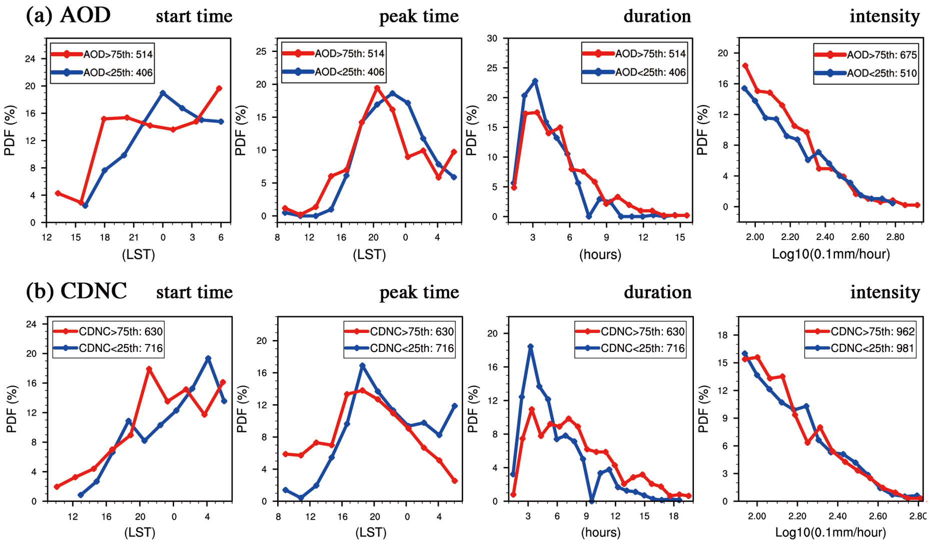


Figure 1. Altitudes (shading, units: m) and selected stations (dots) in the BTH region (red box, 36–41° N, 114–119° E).



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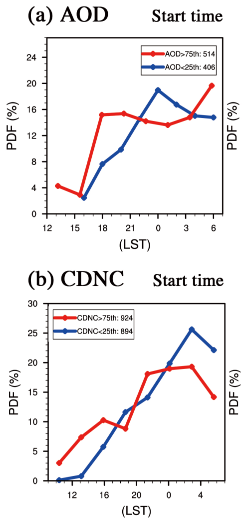

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Figure 2. PDF of start time (units: LST), peak time (units: LST), duration (units: hours) and intensity (units: 0.1mm/hour) of heavy rainfall on selected clean (blue lines) and polluted (red lines) conditions, respectively

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

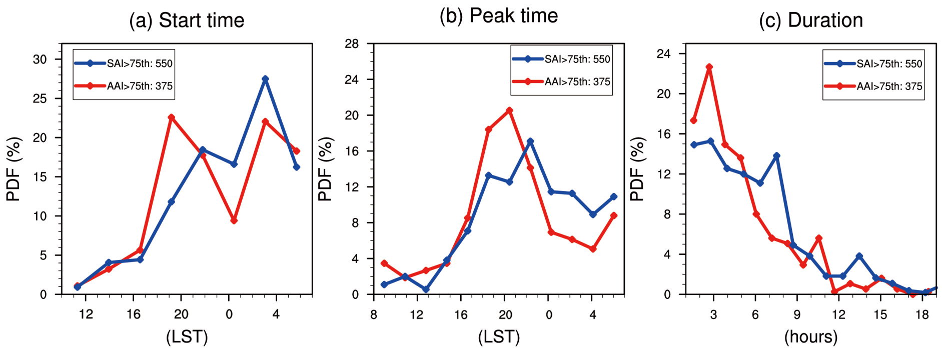

Figure 3. PDF of (a) start time (units: LST), (b) peak time (units: LST), and (c) duration (units: hours) of heavy rainfall on the days that SAI more than 75th tercile (blue lines) and days that AAI more than 75th tercile (red lines), during early summers from 2005 to 2012. The differences between two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

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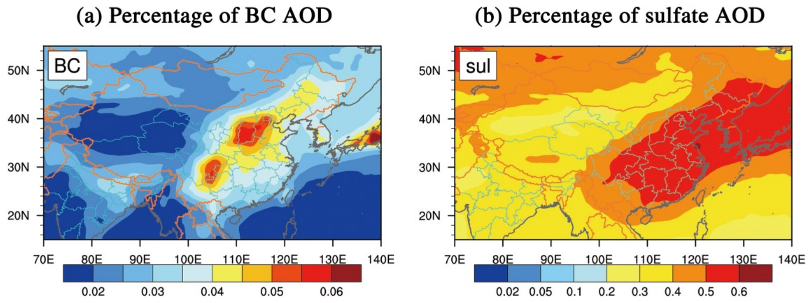

Figure 4. Percentages of AOD for (a) BC and (b) sulfate in JJA during 2002 to 2012.

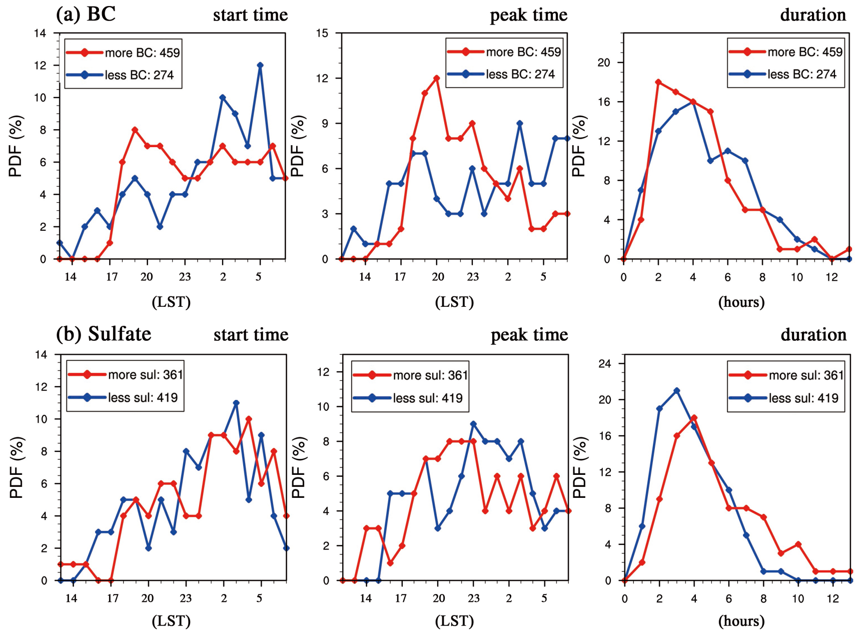

Figure 5. PDF of start time (units: LST), peak time (units: LST) and duration (units: hours) of heavy rainfall in different conditions of (a) BC and (b) sulfate. Blue/red lines stand for the condition of less/more BC or sulfate during early summers from 2003 to 2012. The differences have passed the significant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

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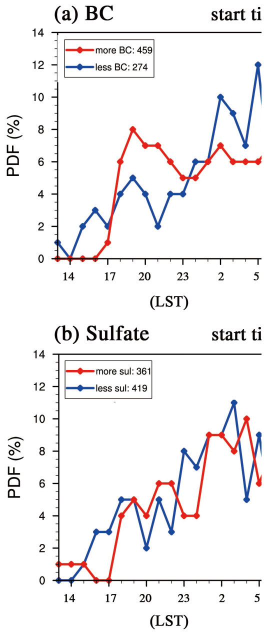

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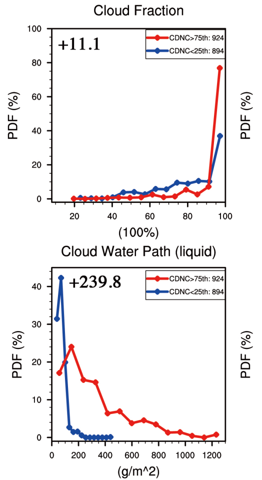

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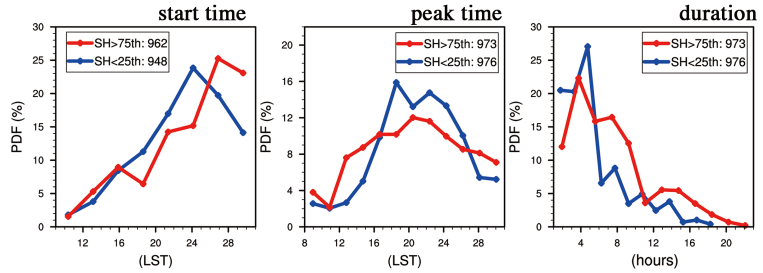
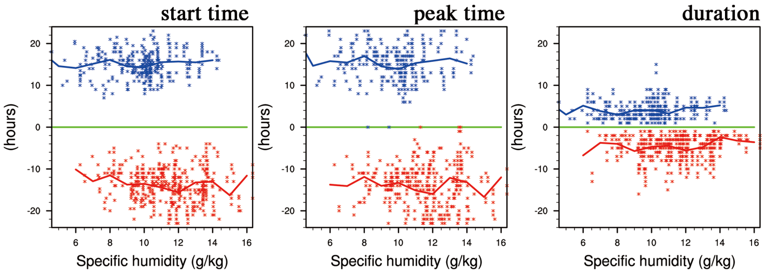
(a) PDF with more/less SH



(b) Scatter distribution using AOD



(c) Scatter distribution using CDNC

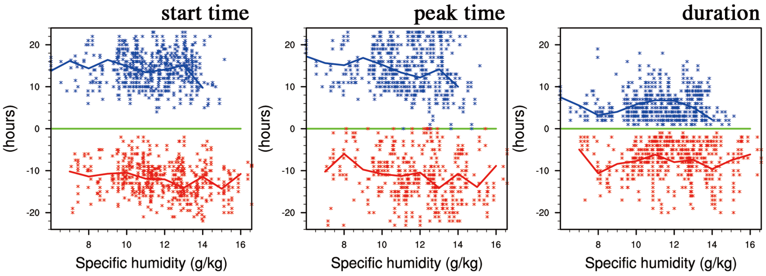


Figure 6. (a) PDF of start time (units: LST), peak time (units: LST), and duration (units: hours) of heavy rainfall in different conditions of less moisture (blue lines, SH at 850 hPa less than 25th tercile) and more moisture (red lines, SH at 850 hPa more than 75th tercile). (b) and (c) are scatter 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 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.

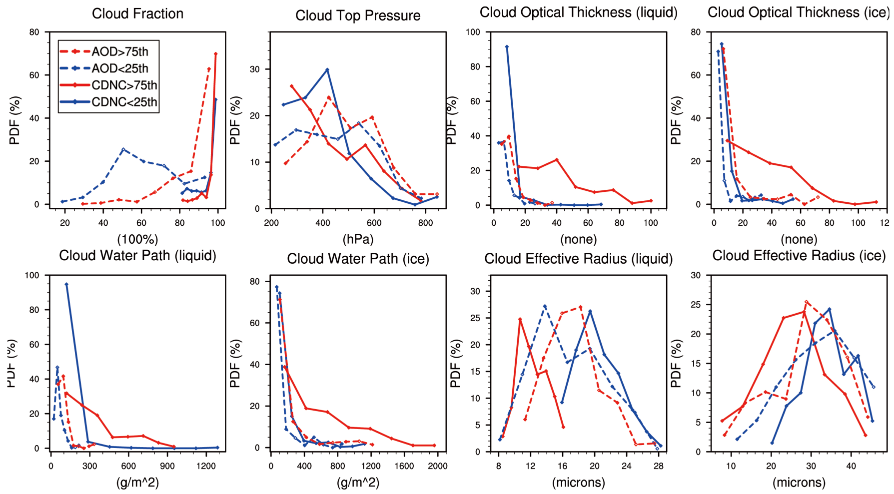


Figure 7. PDF of CF (units: %), CTP (units: hPa), COT (liquid and ice, units: none), CWP (liquid and ice, units: g/m^2) and CER (liquid and ice, units: μm) on selected clean (blue dash lines: AOD<25th tercile; blue solid lines: CDNC<25th tercile) and polluted (red dash lines: AOD>75th tercile; red solid lines: CDNC>75th tercile) heavy rainfall days. The differences between clean and polluted cases have all passed the significant test of 95%.

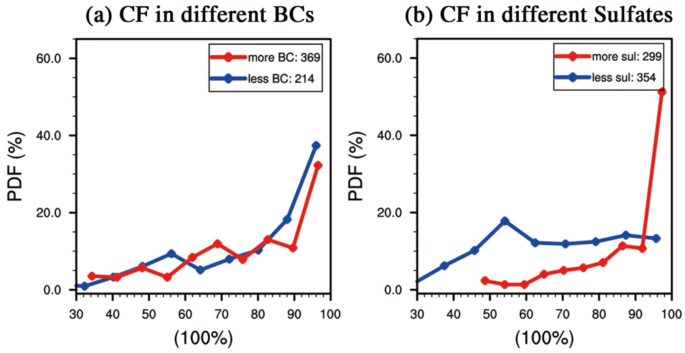


Figure 8. PDF of CF (units: 100%) respectively for selected less BC/sulfate (blue lines) and more BC/sulfate (red lines) cases with heavy rainfall during 10 early summers (2003-2012).

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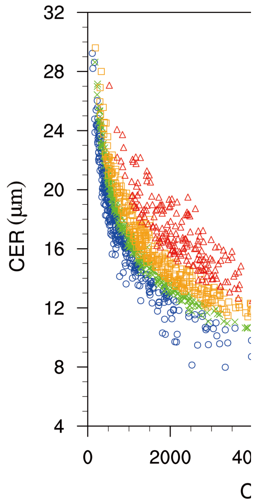

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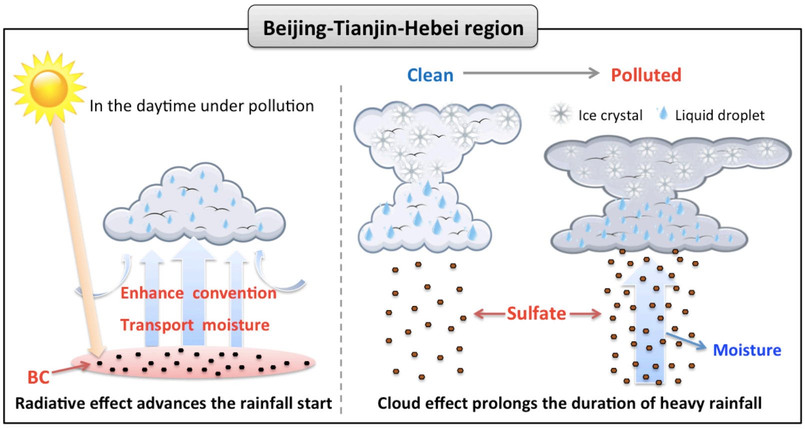

Figure 9. A schematic diagram for aerosols impact on heavy rainfall over Beijing-Tianjin-Hebei region.