

Answers to Referee #1's comments

Thanks a lot for your helpful comments, all of which are well taken. The point-by-point response is given below in black.

Major:

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

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

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

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

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

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

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

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

3. The results using MERRA2 reanalysis data are deleted.

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

Specific comments

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

R: We agree, this part is deleted.

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

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

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

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

15. L300 P9: The authors do try and use BC and SO₄ mass concentrations, which sort of gets around the issues with AOD and RH. The model level that the concentration is taken at is not specified. However, the authors still partition by AOD, and just use BC and SO₄ to partition the high and low AOD cases. The results shown in Fig. 8 are not terribly convincing. It is stated that the results are significant at 95% confidence. What does this mean? Are the means different and 95% confidence or are the values of the PDF in each bin different at 95% confidence? From just looking at the PDFs it seems like only the BC peak time and SO₄ duration are all that are different. The CF PDFs in Fig. 9 look different for SO₄, but the jump in the PDF of CF at 90% does not look terribly robust. This may change if more bins are used in the PDF.

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

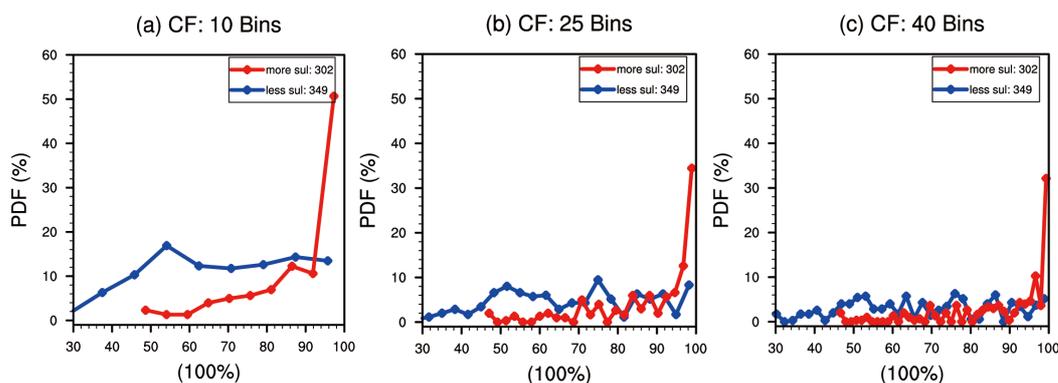


Figure A1. PDF of CF (units: %) in different conditions of sulfate using (a) 10 bins, (b) 25 bins and (c) 40 bins. Blue/red lines stand for the condition of less/more sulfate.

Boucher, O., and Lohmann, U.: The sulfate-CCN-cloud albedo effect, *Tellus B*, 47, 281-300, 10.1034/j.1600-0889.47.issue3.1.x, 1995.

Christensen, M. W., Neubauer, D., Poulsen, C., Thomas, G., McGarragh, G., Povey, A. C., Proud, S., and Grainger, R. G.: Unveiling aerosol-cloud interactions Part 1: Cloud contamination in satellite products enhances the aerosol indirect forcing estimate, *Atmos. Chem. Phys. Discuss.*, 2017, 1-21, 10.5194/acp-2017-450, 2017.

Lowenthal, D. H., Borys, R. D., Choulaton, T. W., Bower, K. N., Flynn, M. J., and Gallagher, M. W.: Parameterization of the cloud dropletsulfate relationship, *Atmos. Environ.*, 38, 287-292, 10.1016/j.atmosenv.2003.09.046, 2004.

- McCarty, W., Coy, L., R. G., A. H., Merkova, D., EB, S., M, S., and K, W.: MERRA-2 Input Observations: Summary and Assessment, Technical Report Series on Global Modeling and Data Assimilation, 46, 2016.
- McCoy, D. T., Bender, F. A. M., Grosvenor, D. P., Mohrmann, J. K., Hartmann, D. L., Wood, R., and Field, P. R.: Predicting decadal trends in cloud droplet number concentration using reanalysis and satellite data, *Atmospheric Chemistry and Physics*, 18, 2035-2047, 10.5194/acp-18-2035-2018, 2018.
- Molod, A., Takacs, L., Suarez, M., and Bacmeister, J.: Development of the GEOS-5 atmospheric general circulation model: evolution from MERRA to MERRA2, *Geosci. Model Dev.*, 8, 1339-1356, 10.5194/gmd-8-1339-2015, 2015.
- Shinozuka, Y., Clarke, A. D., Nenes, A., Jefferson, A., Wood, R., McNaughton, C. S., Ström, J., Tunved, P., Redemann, J., Thornhill, K. L., Moore, R. H., Latham, T. L., Lin, J. J., and Yoon, Y. J.: The relationship between cloud condensation nuclei (CCN) concentration and light extinction of dried particles: indications of underlying aerosol processes and implications for satellite-based CCN estimates, *Atmos. Chem. Phys.*, 15, 7585-7604, 10.5194/acp-15-7585-2015, 2015.
- Twohy, C. H., Coakley, J. A., and Tahnk, W. R.: Effect of changes in relative humidity on aerosol scattering near clouds, *Journal of Geophysical Research: Atmospheres*, 114, n/a-n/a, 10.1029/2008JD010991, 2009.
- Jung, W. S., Panicker, A. S., Lee, D. I., and Park, S. H.: Estimates of aerosol indirect effect from Terra MODIS over Republic of Korea, *Advances in Meteorology*, 2013 (976813): 1-8, <http://dx.doi.org/10.1155/2013/976813>, 2013.
- Qiu, Y., Zhao, C., Guo, J., and Li, J.: 8-Year ground-based observational analysis about the seasonal variation of the aerosol-cloud droplet effective radius relationship at SGP site. *Atmos. Environ.* 164: 139-146, 2017.
- Twomey, S.: The influence of pollution on the shortwave albedo of clouds, *J. Atmos. Sci.*, 34, 1149–1152, doi:10.1175/1520-0469(1977)034<1149:TIOPOT>2.0.CO;2, 1977.

Answers to Referee #2'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.

In this work, the authors look at how properties of gauge-measured rainfall is linked to satellite and reanalysis aerosol and cloud properties. They demonstrate that there are strong correlations between the satellite retrieved aerosol and cloud properties, similar to previous work. They also show a correlation between the timing of the precipitation and the retrieved aerosol. Using reanalysis meteorological and aerosol data, they demonstrate that these relationships between aerosol, cloud and precipitation vary as a function of large-scale humidity and aerosol type.

This work contains several interesting ideas, the use of the gauge precipitation data overcomes issues with the satellite retrieved datasets used in previous studies and the authors have used their knowledge of the local meteorology to attempt to account for meteorological covariations. However, it is not clear that these covariations have actually been accounted for. Unfortunately, this means that many of the inferences in this work may be overstating the role of aerosols. The results in this paper are potentially interesting, but the authors either need to show more definitively that aerosols are driving these relationships or to tone down the assertions that aerosols are the controlling factor. As such, I would only recommend publication after major corrections.

1 Main points

1.1 The role of aerosols

Many previous studies have shown that aerosol optical depth (AOD) is not a good proxy for CCN (Stier, 2016) and is strongly correlated to humidity, which can generate correlations between AOD and cloud fraction (CF; Quaas et al., 2010), as well as other cloud properties (Christensen et al., 2017). It has been shown that using large scale humidity to account for this issue is insufficient (Boucher and Quaas, 2012).

This can even affect studies of cloud development similar to those in this study (e.g. Matsui et al., 2006; Meskhidze et al., 2009; Gryspeerd et al., 2014). This is due to the AOD-CF relationship resulting in different initial cloud distributions for the high and low AOD populations. As cloud development is linked to this initial state, this leads to the AOD-CF relationship (known to be strongly controlled by relative humidity) generating a link between AOD and cloud (or precipitation) development.

As the authors note, it is not easy to isolate the impact of aerosols from that of meteorology in a purely observational study. By restricting the circulation patterns analysed, the authors have gone some way towards doing this, but subsetting by reanalysis humidity alone has been shown by previous studies to be unable to account for the impact of meteorology. This is a difficult task, one that may not be achievable with current data. However, in that case, the conclusions would have to be changed to reflect this.

R: We certainly are in agreement with the issues raised by the reviewer and the manuscript has been completely re-written. To respond the comments, we have done the following to address them. In addition to AOD, two indicators, the retrieved CDNC and ultraviolet AI, are also used in the analysis. CDNC

calculated by COT and CER from MODIS, is used to separate the different CCN conditions and verify the results based on AOD; see Section 2.1.3. AI from OMI is used to identify rainfall days having absorbing aerosols versus scattering aerosols; see Section 2.1.2. The uncertainties associated with these indicators are given in Section 6.3.

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

1.2 Data choices

The MERRA data is not suitable for use as a cloud product, as it is not a measurement, but a model parametrisation. I am not clear to the extent which MERRA represents aerosol-cloud interactions, but if they are not included in the model, the relationship between MERRA clouds (depending only on meteorology) and observed aerosol would be indicative of a meteorological covariation (similar to the results presented in Boucher and Quaas (2012)).

The MODIS 1 by 1 degree CTP is an average of multiple retrievals. In cases with multiple layers of cloud in the same gridbox, the average CTP may be less than 600hPa despite the gridbox containing large amounts of low cloud. The histograms in the MODIS product could be used to better ensure a low contribution of low level cloud.

To account for the impact of humidity on the AOD retrieval, it may be possible to use reanalysis aerosol (McCoy et al., 2017). However, care should be taken in strongly precipitating environments, as this has a quite different spatial sampling compared to MODIS. Whilst MACC/MERRA reanalysis may be able to account for uncertainties in the retrieval, it has quite different relationships to precipitation which should be considered if it is used (Gryspeerd et al., 2015).

R: We agree on the remarks on MERRA's cloud data, and since it is a very minor part of the study, we have eliminated its use completely.

We regard the clouds with the CTP above 600hPa as the mixed-phase clouds, but actually the results of liquid cloud properties in the study (Figure 6 in the revised manuscript) might come from the liquid clouds (low-level clouds) and also the liquid part of mixed phase clouds since we cannot distinguish the liquid part of mixed-phase clouds from the pure liquid clouds in our observation study. We also checked the changes of pure liquid clouds (when the ice properties are missing) as shown in Figure A1, the results are the same as the clouds with CTP above 600hPa. We added sentences to clarify this issue; see Lines 332-336, Page 10.

On AOD data, both MACC and MERRA datasets show very similar features with MODIS (such as the result of MACC in Fig. A2). Since the MACC/MERRA reanalysis data are not completely equal to observational data, we did not address it in the revised manuscript.

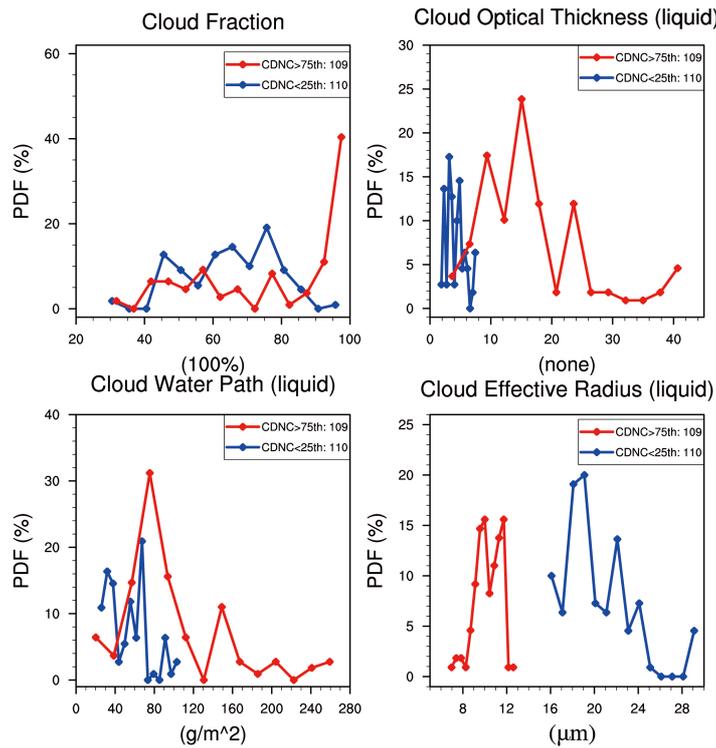


Figure A1. PDF of CF (units: %), COT (units: none), CWP (units: g/m²) and CER (units: μm) for only liquid clouds (when ice COT/CWP/CER is missing) on selected clean (blue lines: CDNC < 25th percentile) and polluted (red lines: CDNC > 75th percentile) heavy rainfall days.

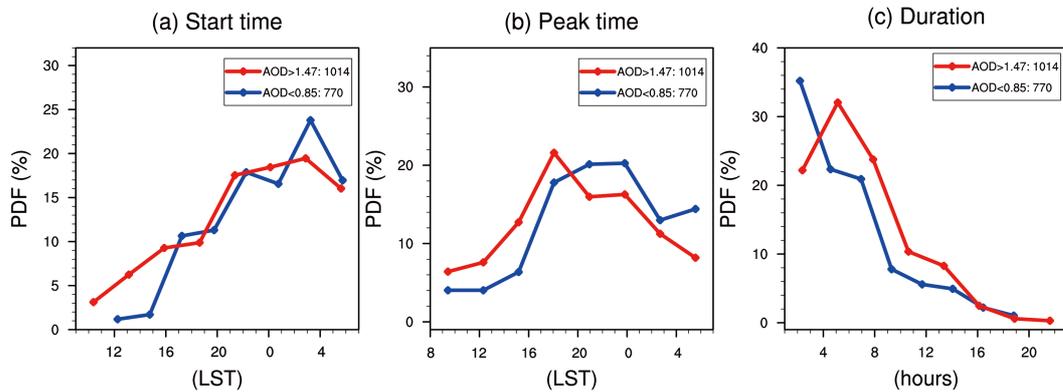


Figure A2. PDF of (a) start time (units: LST), (b) peak time (units: LST), and (c) duration (units: hours) of heavy rainfall on selected clean (blue lines) and polluted (red lines) conditions using AOD from MACC.

1.3 Physical explanations

I found some of the explanations of the aerosol-cloud correlations confusing. In particular, the explanation for the moisture dependence of the correlation between AOD and CTP (L278) is very different from the process described in the references.

The standard impact of aerosol on collision-coalescence (C-C) is to reduce its efficiency (as smaller droplets have a reduced coalescence probability) suppressing precipitation (Albrecht, 1989). However, the explanation in section 4.3 seems to be suggesting that increased aerosol and droplet numbers enhance C-C, making precipitation more likely.

This is not supported by the references (as far as I understand them). There may also be similar explanations, for example - an increase in the low cloud fraction with increasing low level humidity could increase the gridbox mean CTP. The already high cloud fraction in the high AOD cases might limit the impact of this meteorological covariation to the low AOD cases only, explaining the different relationships of humidity, AOD and CTP observed in Fig. 3f.

This is not to say that the authors explanation is wrong, but it should be better supported by references, or with calculations or data if it is a new hypothesis.

R: As discussed in Section 6 in the revised manuscript, the cloud characteristics are much more complex and sensitive to the indicators used. While both AOD and CDNC indicate weaker invigoration effect, the liquid CER shows positive association with AOD but negative with CDNC. Therefore, in the revised manuscript, we only lay out the hypotheses and more insights will have to rely on model simulations.

2 Minor points

L61 - Qian et al. - not in references

R: Added.

L65 - complicity - complicated nature?

R: We have modified it; see Lines 70-72, Page 3.

L69 - The Twomey effect is only the change in droplet number resulting in a change in cloud albedo, not the collective result of all aerosol effects on liquid clouds.

R: Thanks for pointing it out, we have revised it; see Lines 73-75, Page 3.

L82 - Gryspeerdt et al. (2014) showed a link between aerosol and precipitation development with another attempt to account for meteorological covariations.

R: Reference added.

L83 - Similar to this study, these other studies have shown aerosol is correlated with a change in precipitation, not that it causes the change.

R: We agree and modify the writing; see Lines 90-92, Page 3.

L110 – A map of the stations/region used would be useful here

R: Yes, added.

L114 – AOD is not necessarily a good proxy for CCN (e.g. Stier et al, 2016)

R: We agree and added CDNC as another proxy.

L125 - 'we suppose' - could this be checked?

R: As seen from the PDF of heavy rainfall start time in Fig. 2, all the events occur after 10:30 LST which is the overpass time of satellite, i.e., the AOD record we used is either before the precipitation starting in that rainfall day or on the previous day before the rainfall event. We have revised the writing; see Lines 133-136, Page 4.

L133 - assimilation definitely reduces the shortcomings of model simulations, but it is not clear that it 'overcomes' them completely.

R: The sentence is modified; see Line 149, Page 5.

L146 - QA of marginal or higher. Marginal is the lowest retrieval confidence other than 'no confidence'. Why is this choice made - does using a higher confidence level strongly impact the results?

R: Since the AOD records with heavy rainfall are not sufficient, to increase the rainfall sample size we chose the data with marginal or higher confidence. We have tested the result using higher confidence, which is similar but not significant as the result in this study.

L159 - Why is a different reanalysis used for the clouds and the meteorology?

R: For meteorology (wind, temperature, humidity etc.), we used both MERRA2 and ERA-interim reanalysis data for consistency. Since ERA-interim reanalysis data does not have three dimensional cloud variables, we used MERRA2 data to examine the cloud effect. However, the latter was taken out of the revised manuscript because they are simulated rather than observed clouds.

L167 – I am not clear why focusing on this time period better identifies the effect of aerosol

R: The reasons for choosing this period are already given in our earlier study (Zhou et al., 2018). Besides the large-scale dynamics, major consideration is that the period has convective rainfall with heavy pollution. To benefit the readers, we have revised the sentences; see Lines 205-208, Page 7.

L174 - These are very high values of AOD. Brennan et al. (2005) suggested that at AOD>0.6, aerosol is likely to be misclassified as cloud. Might this affect the results here?

R: We have been very careful about this issue. First, there are only a few days with the AOD less than 0.6 over BTH region. And we have tried selecting the samples in which AOD<1.0 to do the same analysis using the percentile method and found the result is similar as shown in Fig. A3. Comparing with AOD <0.57 (which is the 25th percentile in AOD < 1.0), the start time and peak time of heavy rainfall is earlier, and the duration is longer when AOD >0.85 (which is the 75th percentile in AOD < 1.0). Besides, the using of CDNC in the revised manuscript can make up for the uncertainties of AOD, thus we did not address this issue in the article.

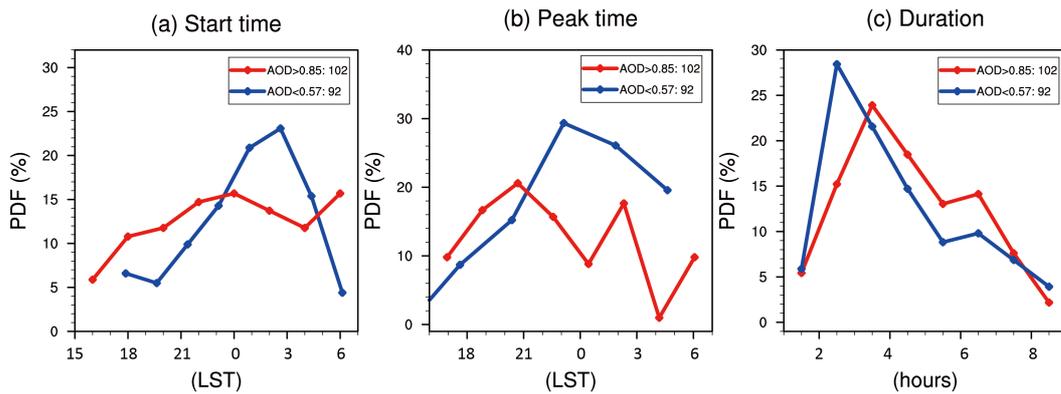


Figure A3. PDF of (a) start time (units: LST), (b) peak time (units: LST), and (c) duration (units: hours) of heavy rainfall on selected clean (blue lines) and polluted (red lines) conditions when AOD less than 1.0.

L190 - There also appears to be a later peak in the precipitation rate at high AOD. Is it clear whether the peak has move earlier or later.

R: Actually there is also a later peak in the PDF of heavy rainfall start time at high AOD, which is at early morning. While in this study, we mainly focus on the heavy rainfall occurred during afternoon and night, since the heavy rainfall occurred in this time is mostly generated by local convection, while the early-morning rainfall might be 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). Besides, the average start time of heavy rainfall in high AOD is in advance in Tab.1 in the manuscript, and the result using CDNC also shows the heavy rainfall moves earlier. A sentence is added to Lines 245-248, Page 8.

L206 - Given that much of the paper is about the development of precipitation, it might be good to point out that the cloud properties are measured at the same time as the aerosol. This is stated in the methods section, but a small reminder would be useful.

R: Thanks. Lines 304-305, Pages 9-10 are added.

L260 - It is not clear what 'nearly 350hPa' means. 340 or 360hPa?

R: This part is totally re-written; see Lines 310-313, Page 10.

L287 - Twomey not Towmey

R: Corrected.

L290 - via enhanced collision-coalescence - this is not the mechanism stated in Yuan et al. 2008, where the positive AOD-effective radius relationship is related to changes in aerosol properties.

R: Yes, it was an incorrect citation. Since we used CDNC instead of AOD to investigate the aerosol indirect effect in the revised version, the results have been changed; see Section 6.1.

L295 - The Wegner-Bergeron-Findeisen process can act whenever supercooled liquid and ice crystals co-exist. As long as liquid droplets exist, it should not depend directly on the supersaturation over liquid,

although if the region is supersaturated with respect to liquid, the liquid droplets can also continue to grow.

R: We are intrigued by the feature that the ice cloud effective radius is decreased when both CDNC and cloud water are increased, and hypothesize that it may be related to the Wegner-Bergeron-Findeisen process. Certainly, modeling studies may provide insights.

L319 - These changes would shift the PDF of CF, but I am not sure it can be said that BC 'corresponds to a slight decrease of CF when CF is more than 90%' as the CF in an aerosol-free atmosphere is not known. Instead, it would be more accurate to use phrases such as 'cloud fractions larger than 90% are less common in high BC environments'.

R: Yes, we have revised the writing; see Lines 415-416, Page 13.

References

- Albrecht, B. A.: Aerosols, Cloud Microphysics, and Fractional Cloudiness, *Science*, 245, 1227–1230, <https://doi.org/10.1126/science.245.4923.1227> , 1989.
- Boucher, O. and Quaas, J.: Water vapour affects both rain and aerosol optical depth, *Nat. Geosci.*, 6, 4–5, <https://doi.org/10.1038/ngeo1692> , 2012.
- Brennan, J., Kaufman, Y., Koren, I., and Rong, L.: Aerosol-cloud interaction-Misclassification of MODIS clouds in heavy aerosol, *IEEE T. Geosci. Remote*, 43, 911–915, <https://doi.org/10.1109/TGRS.2005.844662> , 2005.
- Christensen, M. W., Neubauer, D., Poulsen, C. A., Thomas, G. E., McGarragh, G. R., Povey, A. C., Proud, S. R., and Grainger, R. G.: Unveiling aerosol–cloud interactions – Part 1: Cloud contamination in satellite products enhances the aerosol indirect forcing estimate, *Atmos. Chem. Phys.*, 17, 13 151–13 164, <https://doi.org/10.5194/acp-17-13151-2017> , 2017.
- Grypsperdt, E., Stier, P., and Partridge, D. G.: Links between satellite-retrieved aerosol and precipitation, *Atmos. Chem. Phys.*, 14, 9677–9694, <https://doi.org/10.5194/acp-14-9677-2014>, 2014.
- Grypsperdt, E., Stier, P., White, B. A., and Kipling, Z.: Wet scavenging limits the detection of aerosol effects on precipitation, *Atmos. Chem. Phys.*, 15, 7557–7570, <https://doi.org/10.5194/acp-15-7557-2015> , 2015.
- Matsui, T., Masunaga, H., Kreidenweis, S. M., Pielke, R. A., Tao, W.-K., Chin, M., and Kaufman, Y. J.: Satellite-based assessment of marine low cloud variability associated with aerosol, atmospheric stability, and the diurnal cycle, *J. Geophys. Res.*, 111, 17 204, <https://doi.org/10.1029/2005JD006097>, 2006.
- McCoy, D. T., Bender, F. A.-M., Mohrmann, J. K. C., Hartmann, D. L., Wood, R., and Grosvenor, D. P.: The global aerosol-cloud first indirect effect estimated using MODIS, MERRA, and AeroCom, *J. Geophys. Res.*, 122, 1779–1796, <https://doi.org/10.1002/2016JD026141> , 2017.
- Meskhidze, N., Remer, L. A., Platnick, S., Negrón Juárez, R., Lichtenberger, A. M., and Aiyyer, A. R.: Exploring the differences in cloud properties observed by the Terra and Aqua MODIS Sensors, *Atmos. Chem. Phys.*, 9, 3461–3475, <https://doi.org/10.5194/acp-9-3461-2009> , 2009.

- Quaas, J., Stevens, B., Stier, P., and Lohmann, U.: Interpreting the cloud cover – aerosol optical depth relationship found in satellite data using a general circulation model, *Atmos. Chem. Phys.*, 10, 6129–6135, <https://doi.org/10.5194/acp-10-6129-2010>, 2010.
- Stier, P.: Limitations of passive remote sensing to constrain global cloud condensation nuclei, *Atmos. Chem. Phys.*, 16, 6595–6607, <https://doi.org/10.5194/acp-16-6595-2016>, 2016.
- Ding, Y. H.: Summer monsoon rainfalls in China. *J. Meteor. Soc. Jpn.* 70: 373-396, 1992.
- Higgins, R. W., Yao, Y., Yarosh, E. S., Janowiak, J. E. and Mo, K. C.: Influence of the Great Plains low-level jet on summertime precipitation and moisture transport over the central United States, *J. Climate*, 10, 481-507, 1997.
- Li, H., Cui, X., Zhang, W., and Qiao, L.: Observational and dynamic downscaling analysis of a heavy rainfall event in Beijing, China during the 2008 Olympic Games, *Atmos. Sci. Lett.*, 17, 368-376, 2016.
- Liu, J., Wang, S., Zhang, W., and Wei, X.: Mechanism analysis of a strong convective weather in Hebei Province, *Advances in Marine Science*, 30, 9-16, 2012.
- Wolyn, P. G., and Mckee, T. B.: The mountain plains circulation east of a 2-km-high north south barrier, *Mon. Weather Rev.*, 122, 1490-1508, 1994.
- Wu, R., Wang, B.: Multi-stage onset of the summer monsoon over the western North Pacific. *Clim. Dyn.* 17: 277-289, 2001.
- Yu, R. C., Zhou, T. J., Xiong, A. Y., Zhu, Y. J., Li, J. M.: Diurnal variations of summer precipitation over contiguous China. *Geophys. Res. Lett.* 34: L017041, 2007.
- 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. <https://doi.org/10.1002/joc.5700>, 2018.

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

3
4 Siyuan Zhou¹, Jing Yang^{1,2*}, Wei-Chyung Wang³, Chuanfeng Zhao^{2,4}, Daoyi Gong^{1,2}, Peijun Shi^{1,2}

5
6 ¹ Academy of Disaster Reduction and Emergency Management, Faculty of Geographical Science, Beijing
7 Normal University, China

8 ² State Key Laboratory of Earth Surface Process and Resource Ecology, Beijing Normal University, China
9 ³ Atmospheric Sciences Research Center, State University of New York, Albany, New York 12203, USA

10 ⁴ College of Global Change and Earth System Science, Beijing Normal University, China

11
12
13 Submitted to ACP
14 Oct 2018

15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32 *Correspondence to: Jing Yang, State Key Laboratory of Earth Surface Process and Resource
33 Ecology/Academy of Disaster Reduction and Emergency Management, Faculty of Geographical Science,
34 Beijing Normal University, 19#Xinjiekouwai Street, Haidian District, Beijing 100875, China. E-mail:
35 yangjing@bnu.edu.cn

Siyuan Zhou 19/2/22 5:15 PM

样式定义: 普通(网站)

Siyuan Zhou 19/2/22 5:15 PM

已删除: Diurnal

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 字体:13 pt

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 字体:13 pt

Siyuan Zhou 19/2/22 5:15 PM

已删除: the

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 字体:13 pt

Siyuan Zhou 19/2/22 5:15 PM

已删除: region: Role of aerosol cloud effect and its sensitivity to moisture

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 字体:13 pt

Siyuan Zhou 19/2/22 5:15 PM

已删除: ³, Wei-Chyung Wang³

Siyuan Zhou 19/2/22 5:15 PM

已删除: ³

Siyuan Zhou 19/2/22 5:15 PM

已删除: ⁴Atmospheric Sciences Research Center, State University of New York, Albany, New York 12203, USA

Siyuan Zhou 19/2/22 5:15 PM

已删除: .

46 **Abstract:** Our recent observational study found that the rainfall diurnal variation over Beijing-Tianjin-Hebei
47 shows distinct signature of the effects of pollutants. Here we used the hourly rainfall measurements together
48 with daily satellite-based information of aerosols and clouds to further study the responses of heavy rainfall
49 and cloud properties to increases of pollutants. While the aerosol optical depth (AOD) and cloud droplet
50 number concentration (CDNC) are both used as pollution indicators to provide a different perspective in
51 rainfall study, CDNC is used exclusively on cloud changes. It is found that both indicators yield three
52 consistent and distinguished responses of heavy rainfall: earlier start time, earlier peak time, and longer
53 duration. However, quantitative differences exist between the two: for the first two responses, the advances
54 are 0.7 and 1.0 hours respectively with AOD, but 2.1 and 4.2 hours respectively with CDNC; the third is
55 prolonged by 0.8 hours with AOD and 2.4 hours with CDNC. In-depth analysis suggests that earlier in both
56 start time and peak time occur in the presence of absorbing aerosols while the longer duration is attributed to
57 scattering aerosols. Changes in cloud statistics caused by aerosols show increases in cloud fraction (11.1%),
58 cloud top pressure (37.8 hPa), the liquid/ice cloud optical thickness (32.2/26.0) and cloud water path
59 (239.8/422.0 g/m³); and decreases in liquid/ice cloud effective radius (8.6/8.7μm). Analyses also indicate that
60 increased moisture tends to decrease the cloud top pressure and enlarge the liquid cloud effective radius,
61 which partially compensate the aerosol effects. Finally, the mechanisms accounted for the aerosol effects on
62 heavy rainfall are hypothesized.

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

64

65 1. Introduction

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

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

Siyuan Zhou 19/2/22 5:15 PM

已删除: study found that, during 2002-2012, the diurnal variation of heavy rainfall over Beijing-Tianjin-Hebei (BTH) region exhibits different characteristics between clean and polluted environment. Here we use satellite cloud-products together with meteorology and aerosol data to further examine the aerosol impact on the associated clouds focusing on its sensitivity to moisture. During the days with large aerosol loading, the characteristics of earlier starting time, earlier peak hour and the longer duration of heavy rainfall are usually accompanied by increased cloud fraction, reduced cloud top height and increased/reduced liquid/ice effective radius. However, the aerosol effects on the cloud top and liquid effective radius are distinct at lower and higher humidity. Different from the radiative effect that black carbon heats the lower troposphere and may generate the earlier start of heavy rainfall, the aerosol cloud effect enhances the efficiency of precipitation and advances the rainfall peak, which may be ascribed to increased cloud droplet number and cloud water, enhanced collision-coalescence and accelerated rainfall formation when the background moisture supply is sufficient. The speculation warrants further numerical experiment to verify.

Siyuan Zhou 19/2/22 5:15 PM

已删除: the depth of the mixed-phase

Siyuan Zhou 19/2/22 5:15 PM

已删除: aerosols impact

Siyuan Zhou 19/2/22 5:15 PM

已删除: has

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 首行缩进: 0 字符

Siyuan Zhou 19/2/22 5:15 PM

已删除: the

113 clouds and mixed-phase clouds (e.g. Guo et al., 2015). Because the aerosol-cloud interactions in different
114 types of clouds are distinct (Gryspeerd et al., 2014), aerosol indirect effect during heavy rainfall is more
115 complicated than its direct effect (Sassen et al., 1995; Sherwood, 2002; Jiang et al., 2008, Tao et al., 2012).
116 For warm clouds, by serving as CCN for more cloud droplets, aerosols can increase cloud albedo so called albedo
117 effect or Twomey effect (Twomey, 1977), lengthen the cloud lifetime so called lifetime effect (Albrecht, 1989), and
118 enhance thin cloud thermal emissivity so called thermal emissivity effect (Garrett and Zhao, 2006). The above
119 effects tend to increase the cloud microphysical stability and suppress warm-rain processes (Albrecht 1989;
120 Rosenfeld et al. 2014). For cold clouds and mixed-phase clouds, many studies reported that the cloud liquid
121 accumulated by aerosols is converted to ice hydrometeors above the freezing level, which invigorates deep
122 convective clouds and intensifies heavy precipitation so called invigoration effect (Rosenfeld and Woodley,
123 2000; Rosenfeld et al., 2008; Lee et al. 2009; Guo et al. 2014). The Twomey effect infers that aerosols serving
124 as CCN increasing the cloud droplets could reduce cloud droplet size within a constant liquid water path
125 (Twomey, 1977). However, the opposite results of relationship between aerosols and cloud droplet effective
126 radius were reported in observations (Yuan et al., 2008; Panicker et al., 2010; Jung et al., 2013; Harikishan et
127 al., 2016; Qiu et al., 2017), which might be related with the moisture supply near the cloud base (Yuan et al.,
128 2008; Qiu et al., 2017). Besides, the influence of aerosols on ice clouds also depends upon the amount of
129 moisture supply (Jiang et al., 2008). Therefore, how the aerosols modify the clouds associated with heavy
130 convective rainfall does not reach a consensus, particularly if considering the different moisture conditions.

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

Siyuan Zhou 19/2/22 5:15 PM
已删除: Due to...ecause the ... [1]

Siyuan Zhou 19/2/22 5:15 PM
已设置格式 ... [2]

Siyuan Zhou 19/2/22 5:15 PM
已删除: increase ... [3]

Siyuan Zhou 19/2/22 5:15 PM
已设置格式 ... [3]

Siyuan Zhou 19/2/22 5:15 PM
已删除:), which were collectively known as Twomey effect. Twomey effect increases cloud The above effects tend to in ... [4]

Siyuan Zhou 19/2/22 5:15 PM
已删除: one day.... few hours, thus s ... [5]

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

- Siyuan Zhou 19/2/22 5:15 PM
已删除: days.
- Siyuan Zhou 19/2/22 5:15 PM
已删除: effect
- Siyuan Zhou 19/2/22 5:15 PM
已删除: . Section 5 discusses
- Siyuan Zhou 19/2/22 5:15 PM
已删除: the
- Siyuan Zhou 19/2/22 5:15 PM
已删除: of aerosols that play on diurnal variation of
- Siyuan Zhou 19/2/22 5:15 PM
已删除: 6
- Siyuan Zhou 19/2/22 5:15 PM
已删除: aerosol
- Siyuan Zhou 19/2/22 5:15 PM
已删除: cloud
- Siyuan Zhou 19/2/22 5:15 PM
已删除: plain
- Siyuan Zhou 19/2/22 5:15 PM
已删除: in BTH region
- Siyuan Zhou 19/2/22 5:15 PM
已删除: similar
- Siyuan Zhou 19/2/22 5:15 PM
已删除: effect
- Siyuan Zhou 19/2/22 5:15 PM
已删除: Aerosol optical depth (
- Siyuan Zhou 19/2/22 5:15 PM
已删除:), which
- Siyuan Zhou 19/2/22 5:15 PM
已删除: an indicator
- Siyuan Zhou 19/2/22 5:15 PM
已删除: the
- Siyuan Zhou 19/2/22 5:15 PM
已删除: The Collection 6 aerosol dataset is created from three separate retrieval algorithms that operate over different surface types: the two "Dark Target" (DT) algorithms for retrieving (1) over ocean (dark in visible and longer wavelengths) and (2) over vegetated/dark-soiled land (dark in the visible), plus the "Deep Blue" (DB) algorithm (... [6])
- Siyuan Zhou 19/2/22 5:15 PM
已删除: we suppose
- Siyuan Zhou 19/2/22 5:15 PM
已删除: most
- Siyuan Zhou 19/2/22 5:15 PM
已删除: since
- Siyuan Zhou 19/2/22 5:15 PM
已删除: starting time
- Siyuan Zhou 19/2/22 5:15 PM
已删除: is mostly after 12:00 LST (Fig. 1).
- Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 删除线

205 2. Data and methodology

206 2.1 Data

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

209 2.1.1 Precipitation data

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

220 2.1.2 Aerosol data

221 AOD is a proxy for the optical amount of aerosol particles in a column of the atmosphere and serves as one of
222 indicators for the division of aerosol pollution condition in this study, was obtained from MODIS (Moderate
223 Resolution Imaging Spectroradiometer) Collection 6 L3 aerosol product with the horizontal resolution of
224 $1^\circ \times 1^\circ$ onboard the Terra satellite (Tao et al., 2015). The quality assurance of marginal or higher confidence
225 was used in this study. The reported uncertainty in MODIS AOD data is on the order of (-0.02-10%),
226 (+0.04+10%) (Levy et al., 2013). The Terra satellite overpass time at the equator is around 10:30 local solar
227 time in the daytime, which is before the occurrence of heavy rainfall events in this study as shown in Fig. 2.
228 Therefore, the AOD used here represents the situation of the air quality in advance of heavy rainfall
229 appearance.

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

275 AI is a method of detecting absorbing aerosols from satellite measurements in the near-ultraviolet wavelength
 276 region (Torres et al., 1998). The positive values of ultraviolet AI are attributed to the absorbing aerosols such
 277 as smoke and dust while the negative values of AI stand for the non-absorbing aerosols (scattering aerosols)
 278 such as sulfate and sea salt (Tariq and Ali, 2015). The near-zero values of AI occur when clouds and Rayleigh
 279 scattering dominate (Hammer et al., 2018). The horizontal resolution of AI data is 1°×1° and it covers the
 280 period of 2005 to 2012.

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

288 2.1.3 Cloud data

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

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

$$308 \text{CDNC} = \frac{C_W^{1/2}}{k} \frac{10^{1/2}}{4\pi\rho_w^{1/2}} \frac{\tau^{1/2}}{R_e^{5/2}} \quad (1)$$

- Siyuan Zhou 19/2/22 5:15 PM
已删除: provided
- Siyuan Zhou 19/2/22 5:15 PM
已删除: which assimilates total AOD retrieved by MODIS to correct for model departures from observed aerosols (Benedetti et al., 2009),
- Siyuan Zhou 19/2/22 5:15 PM
已删除: two-dimensional AOD and three-dimensional aerosol mass concentration
- Siyuan Zhou 19/2/22 5:15 PM
已删除: overcome
- Siyuan Zhou 19/2/22 5:15 PM
已删除: shortcoming
- Siyuan Zhou 19/2/22 5:15 PM
已删除: .
- Siyuan Zhou 19/2/22 5:15 PM
已删除: and
- Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 字体颜色: 自动
- Siyuan Zhou 19/2/22 5:15 PM
已删除: vertical resolution is 60 levels
- Siyuan Zhou 19/2/22 5:15 PM
已删除: ., of which the time interval is six-hour.
- Siyuan Zhou 19/2/22 5:15 PM
已删除: estimates using aircraft
- Siyuan Zhou 19/2/22 5:15 PM
已删除: .
- Siyuan Zhou 19/2/22 5:15 PM
已删除: (MAS)
- Siyuan Zhou 19/2/22 5:15 PM
已删除: Likewise, the quality assurance of marginal or higher confidence was used in this study. [7]
- Siyuan Zhou 19/2/22 5:15 PM
已删除: the

331 Where C_w is the moist adiabatic condensate coefficient, and its value depends slightly on the temperature of
332 the cloud layer, ranging from 1 to $2.5 \times 10^{-3} \text{ gm}^{-4}$ for a temperature between 0 °C and 40 °C (Brenguier,
333 1991). In this study, we calculated the C_w through the function of the temperature (see Fig.1 in Zhu et al.,
334 2018) at a given pressure that is 850 hPa. And we have tested the sensitivity of CDNC to the amount of C_w
335 and found it almost keeps the same when the C_w changes from 1 to $2.5 \times 10^{-3} \text{ gm}^{-4}$. The coefficient k is the
336 ratio between the volume mean radius and the effective radius and varies between 0.5 and 1 (Brenguier et al.,
337 2000). Here we used $k = 1$ for that we cannot get the accurate value of k and the value of k does not influence
338 the rank of CDNC for the division of aerosol condition in this study. ρ_w is cloud water density. τ and Re are
339 the COT and CER obtained from MODIS Collection 6 L3 cloud product onboard the Terra satellite.

340 **2.1.4 Other meteorological data**

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

$$347 \text{AH} = \frac{1000e}{R_v T} \quad (2)$$

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

$$350 q = \frac{0.622e}{P - (1 - 0.622)e} \quad (3)$$

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

354 **2.2 Methodology**

355 **2.2.1 Method of interpolation**

356 We used both station data of gauge-based precipitation and gridded data including aerosols, clouds and other
357 meteorological variables. Gridded datasets in this study were downloaded with the horizontal resolution of
358 $1^\circ \times 1^\circ$, which are consistent with the resolution of MODIS L3 product. To unify the datasets, we interpolated
359 all the gridded datasets onto the selected 176 rainfall stations using the average value in a $1^\circ \times 1^\circ$ grid as the
360 background condition of each rainfall station, i.e., the stations in the same $1^\circ \times 1^\circ$ grid have the same aerosol,
361 cloud and meteorological conditions.

Siyuan Zhou 19/2/22 5:15 PM

已删除: the

Siyuan Zhou 19/2/22 5:15 PM

已删除: relative

Siyuan Zhou 19/2/22 5:15 PM

已删除: (RH),

Siyuan Zhou 19/2/22 5:15 PM

已删除: To unify the datasets, we interpolated the gridded datasets into

367 **2.2.2 Selection of sub-season and circulation**

368 Consistent with our previous work, we focused on early summer (1 June to 20 July) before the large-scale
369 rainy season starts, in order to remove the large-scale circulation influence and identify the effect of aerosols on
370 local convective precipitation, because BTH rainfall during this period is mostly convective rainfall (Yu et al.,
371 2007) with heavy pollution (Zhou et al., 2018). And to unify the background atmospheric circulation, we only
372 selected the rainfall days with southwesterly flow, which is the dominant circulation accounting for 40% of
373 total circulation patterns over the BTH region during early summer (Zhou et al., 2018).

374 **2.2.3 Classification of the heavy rainfall, clean/polluted and moisture conditions**

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

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

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

394 **2.2.4 Statistical analysis**

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

Siyuan Zhou 19/2/22 5:15 PM

已删除: ... [8]

Siyuan Zhou 19/2/22 5:15 PM

已删除: (Zhou et al., 2018),... we f... [9]

Siyuan Zhou 19/2/22 5:15 PM

已删除: 2... Classification of the h... [10]

Siyuan Zhou 19/2/22 5:15 PM

已删除: samples...ays when the ho... [11]

Siyuan Zhou 19/2/22 5:15 PM

已删除: Using the same percentile

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 首行缩进: 0 cm, 不调整西文与中文之间的空格, 不调整中文和数字之间的空格

Siyuan Zhou 19/2/22 5:15 PM

已删除: with southwesterly

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 字体颜色: 自动

Siyuan Zhou 19/2/22 5:15 PM

已删除: 3

Siyuan Zhou 19/2/22 5:15 PM

已删除: clean and pollution days or... [12]

440

441 3 Distinct characteristics of heavy rainfall diurnal variation associated with aerosol pollution

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

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

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

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

473

- Siyuan Zhou 19/2/22 5:15 PM
已删除: days
- Siyuan Zhou 19/2/22 5:15 PM
已删除: The
- Siyuan Zhou 19/2/22 5:15 PM
已删除: time
- Siyuan Zhou 19/2/22 5:15 PM
已删除: lb).
- Siyuan Zhou 19/2/22 5:15 PM
已删除: change
- Siyuan Zhou 19/2/22 5:15 PM
已删除: starting
- Siyuan Zhou 19/2/22 5:15 PM
已删除: starting
- Siyuan Zhou 19/2/22 5:15 PM
已删除: for the
- Siyuan Zhou 19/2/22 5:15 PM
已删除: of the starting time
- Siyuan Zhou 19/2/22 5:15 PM
已删除: lb
- Siyuan Zhou 19/2/22 5:15 PM
已删除: of heavy rainfall
- Siyuan Zhou 19/2/22 5:15 PM
已删除: nearly
- Siyuan Zhou 19/2/22 5:15 PM
已删除: .
- Siyuan Zhou 19/2/22 5:15 PM
已删除: lc,
- Siyuan Zhou 19/2/22 5:15 PM
已删除: on the polluted days
- Siyuan Zhou 19/2/22 5:15 PM
已删除: Therefore, the following only focuses on investigating why the starting time, peak time and duration of heavy rainfall change with pollution in diurnal time scale.

493
494
495
496
497
498
499
500
501
502
503
504
505
506
507
508
509
510
511
512
513
514
515
516
517
518
519
520
521
522
523
524
525
526

4 Impacts of different aerosols on rainfall diurnal variation

Using the indicator of AI, we further investigate the different changes of rainfall characteristics 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. The rainfall start time on absorbing aerosol days shows a significant advance with the maximum frequency occurring at 20:00 LST, compared with the 3:00 LST on scattering aerosol days (Fig. 3a). Similarly, the rainfall peak time also shows earlier on absorbing aerosol days, with an average advance of 1.7 hours (Fig. 3b). The rainfall duration on scattering aerosol days shows longer than that on absorbing aerosol days, which are 5.9 hours and 5.0 hours respectively in average. All the differences above 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 effect on heavy rainfall that absorbing aerosols may generate the heavy rainfall in advance and the scattering aerosols may delay and prolong the heavy rainfall.

To further distinguish the effects of the absorbing/scattering aerosols on the heavy rainfall, we purposely re-examine the above findings through BC/sulfate that can represent typical absorbing/scattering aerosols over 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 in 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 the role of them in 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 were shown for the higher and lower BC cases in Fig. 5a, respectively. The most striking result is that the maximum frequency of rainfall start time in high BC cases evidently shifts earlier by 7 hours from 19:00 LST to 2:00 LST. Meanwhile, compared with low BC cases, the mean peak time in high BC cases shows 1.0 hour earlier than that in low BC cases. And the duration of heavy rainfall is slightly shorter in high BC cases with the mean difference of 0.2 hours. These features of higher 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. The behaviors of higher sulfate cases exhibit similar with the above scattering aerosol effect (Fig. 5b).

5 Cloud effect of aerosols with inclusion of moisture

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

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

Siyuan Zhou 19/2/22 5:15 PM
已移动(插入) [1]

Siyuan Zhou 19/2/22 5:15 PM
已移动(插入) [2]

Siyuan Zhou 19/2/22 5:15 PM
已移动(插入) [3]

Siyuan Zhou 19/2/22 5:15 PM
已移动(插入) [4]

Siyuan Zhou 19/2/22 5:15 PM
已删除: ... [13]

Siyuan Zhou 19/2/22 5:15 PM
已删除: days

Siyuan Zhou 19/2/22 5:15 PM
已删除: on

Siyuan Zhou 19/2/22 5:15 PM
已删除: diurnal variation

Siyuan Zhou 19/2/22 5:15 PM
已删除: associated cloud features on clean and polluted days. The differences of

Siyuan Zhou 19/2/22 5:15 PM
已删除: diurnal

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

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

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

560 5.2 Influence of CCN and moisture on cloud properties

561 The different moisture supply under the cloud base can influence the cloud properties as well as the effect of
562 aerosols on cloud properties (Yuan et al., 2008; Jiang et al., 2008; Jung et al., 2013; Qiu et al., 2017). It is hard
563 to completely remove the moisture effect on the above results in a pure observational study. Since the
564 southwesterly circulation cannot only transport pollutants but also moisture to the BTH region (Wu et al.,
565 2017), more pollution usually corresponds to more moisture (Sun et al., 2015). Because the moisture supply
566 for BTH is mainly transported via low-level southwesterly circulation, we purposely used the AH at 850 hPa
567 as the indicator of moisture condition. To identify the effect of aerosols on clouds and its sensitivity to
568 moisture, we purposely investigated the changes of above cloud properties with different conditions of CDNC

Siyuan Zhou 19/2/22 5:15 PM
已删除: circumstances...ondition b... [14]

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 首行缩进: 0 字符
Siyuan Zhou 19/2/22 5:15 PM
已删除: 2.... Both liquid and ice CC... [15]

Siyuan Zhou 19/2/22 5:15 PM
已上移 [1]: 3a).

Siyuan Zhou 19/2/22 5:15 PM
已删除: The major characteristics are that the peak of liquid water occurs in the evening (at 20:00-23:00 LST) (Fig. 3a) while the ice water appears in the mid-night (at 20:00-3:00 LST) (Fig. 3d). Compared with clean condition, the amount of the liquid and ice water are both significantly increased on polluted days. Meanwhile, the peak value of liquid water appears much earlier by almost 8 hours than that on clean days. i.e., the peak of the liquid water occurs at 14:00 LST under pollution (Fig.

Siyuan Zhou 19/2/22 5:15 PM
已上移 [2]: 3b).

Siyuan Zhou 19/2/22 5:15 PM
已删除: The ice water exhibits the similar shift of its peak under pollution and its maximum center appears in the afternoon (at 14:00-17:00 LST) rather than the mid-night (Fig. 3e). The difference of ice water between polluted and clean condition also indicates that the cloud top on polluted days is lower than that on clean days (Fig. 3f), which is consistent with the result in Fig. 2

Siyuan Zhou 19/2/22 5:15 PM
已删除: aerosols correspond...DNC... [16]

Siyuan Zhou 19/2/22 5:15 PM
已删除: 4...2 Changes...influence... [17]

Siyuan Zhou 19/2/22 5:15 PM
已删除: condition...upply under the... [18]

682 and moisture, respectively. We categorized all cases of heavy rainfall into four groups, which are (1) clean and
683 dry, (2) polluted and dry, (3) clean and wet, (4) polluted and wet, and checked the changes of above cloud
684 properties, as shown in Tab. 3. Here “clean/polluted” refers to the CDNC on that rainfall day less/more than
685 25th/75th percentile of the CDNC among the whole rainfall days, and similarly, the “dry/wet” refers to the AH
686 on that rainfall day less/more than 25th/75th percentile of itself among the whole rainfall days. We made the
687 significant test of differences between group 1 and 2, group 1 and 3, group 2 and 4, group 3 and 4.

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

698 Comparing the results of group 1 and 3, we can get the changes of cloud properties related only to moisture
699 on the same clean condition. A common feature is that CF, cloud top height, COT and CWP both for liquid
700 and ice clouds exhibit increases along with the increase of AH (the decrease of CTP corresponds to the
701 increase of cloud top height). Compared with the CF on clean and dry condition (group 1), the increase of CF
702 on clean and wet condition (group 3) is larger than that on polluted and dry condition (group 2), which
703 indicates the influence of moisture on CF might be larger than the influence of CCN. In contrast with CF, the
704 increases of COT and CWP both for liquid and ice clouds in group 2 are 2-3 times larger than that in group 3,
705 which indicates that the influences of moisture on COT and CWP are evidently smaller than the influence of
706 CCN. The influences of moisture on liquid and ice CER are not significant on the same clean condition. On
707 polluted condition, comparing group 2 and 4, we found the same changes are the increase of CF, liquid COT
708 and CWP, and the decrease of CTP, while the influences of moisture on ice COT and CWP on the polluted
709 condition become not significant. When the moisture increases, the liquid CER on polluted condition is
710 increased and the ice CER is decreased.

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

Siyuan Zhou 19/2/22 5:15 PM
已删除: change
Siyuan Zhou 19/2/22 5:15 PM
已删除: on the
Siyuan Zhou 19/2/22 5:15 PM
已删除: days and
Siyuan Zhou 19/2/22 5:15 PM
已删除: days (Fig. 5).

Siyuan Zhou 19/2/22 5:15 PM
已删除: A common feature is that all examined variables of clouds exhibit increases along with the increase of moisture on both clean and polluted days (Fig. 5). If fixing the moisture, the amounts of CF, COT (both liquid and ice), CWP (both liquid and ice) become larger on the polluted days, which are consistent with the above-mentioned results without removing the moisture effect in Sect. 4.1. However, the aerosol effect on CTP is evidently distinct between low and high RH conditions (Fig. 5f). When the RH is relatively low (<70%), the amount of CTP on polluted days is larger than that on clean days. In contrast, the CTP becomes smaller when the RH is relatively high (>70%). That is to say, aerosols reduce the cloud top at lower RH but increase it at higher RH. ... [19]

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

747

748 **6 Discussion**

749 **6.1 Possible effect of aerosols on cloud with inclusion of moisture**

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

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

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

Siyuan Zhou 19/2/22 5:15 PM

已删除: . However, why the aerosol effect on cloud top and liquid CER depends on different moisture conditions has not been clarified yet.

Siyuan Zhou 19/2/22 5:15 PM

已删除: In terms of cloud top, we speculate the following mechanisms in clean and polluted condition. On the clean days with fewer moisture, the fewer cloud droplets cause the delayed precipitation due to relatively depressed collision-coalescence process, thus the clouds tend to develop vertically to a higher altitude, which also corresponds to the delayed formation of ice clouds (Fig. 3d). On the polluted days, the increased aerosols (CCN) can increase the cloud droplet number (Squires and Twomey, 1966), which can enhance the collision-coalescence process (Rosenfeld, 1999; Liu et al., 2003). When the moisture supply is sufficient, the cloud drops can become larger via adequate collision-coalescence and easily convert to rain drops, which facilitates the advance of rainfall start. After the rainfall started, the cloud top is restricted to grow higher. Therefore, the cloud top exhibits relatively lower in polluted cases over BTH region (Fig. 3f). ... [20]

Siyuan Zhou 19/2/22 5:15 PM

已删除: but

Siyuan Zhou 19/2/22 5:15 PM

已删除: .

Siyuan Zhou 19/2/22 5:15 PM

已删除: So far

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

807 6.2 Different roles of aerosol radiative effect and cloud effect in heavy rainfall

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

820 The delayed start of heavy rainfall with higher scattering aerosols in Fig. 2a and higher sulfate in Fig. 4b is
821 consistent with many studies that both the radiative effect and cloud effect of sulfate-like aerosols could delay
822 or suppress the occurrence of rainfall (Guo et al., 2013; Wang et al., 2016; Rosenfeld et al. 2014). Sulfate-like
823 aerosols as scattering aerosols could prevent the shortwave radiation from arriving at the surface thus cool the
824 surface and stabilize the atmosphere, which suppresses the rainfall formation (Guo et al., 2013; Wang et al.,
825 2016). Sulfate-like aerosols serving as CCN can also suppress the rainfall by cloud effect through reducing the
826 cloud droplet size and thus suppressing the collision-coalescence process of cloud droplets (Albrecht 1989;
827 Rosenfeld et al. 2014). Figure 8b does shows that in contrast with BC, the CF larger than 90% is significantly
828 increased in the high sulfate environment, which indicates the sulfate-like aerosols have evident influence on
829 the clouds. We also verified that the cloud droplet shifts to a smaller size when the CDNC increases (Fig. 6) in
830 Sect. 5, indicating that the cloud effect of aerosols could lead to the delay of the heavy rainfall occurrence.
831 Another significant feature is the longer duration of heavy rainfall in the high scattering aerosol cases and high
832 sulfate cases (Fig 3c&5b). We speculate that the longer duration is caused by the cloud effect of sulfate-like
833 aerosols. When CCN increases over BTH region, the cloud droplet size is decreased but the cloud water is
834 increased (Fig. 6). Therefore, the rainfall start time is delayed for the reduced collision-coalescence of cloud
835 droplets, while the duration might be prolonged due to the significant increase of cloud water. To further
836 investigate the mechanism of longer duration, we need the assistance of numerical model simulations in the
837 future work.

Siyuan Zhou 19/2/22 5:15 PM
已删除: explore

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 字体:非 加粗

Siyuan Zhou 19/2/22 5:15 PM
已删除: 5 Aerosol

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 无

Siyuan Zhou 19/2/22 5:15 PM
已删除: on...n heavy rainfall diur... [21]

Siyuan Zhou 19/2/22 5:15 PM
已删除: Our previous study has indicated that the radiative effect of BC low-level warming may facilitate the convective rainfall generation (Zhou et al., 2018). Based on the changes of cloud properties address... [22]

Siyuan Zhou 19/2/22 5:15 PM
已上移 [3]: (Gunthe et al., 2011).

Siyuan Zhou 19/2/22 5:15 PM
已删除: We purposely selected the cases with different BC/sulfate concentrations to compare the role of BC/sulfate on the diurnal variation of the heavy rainfall.

Siyuan Zhou 19/2/22 5:15 PM
已上移 [4]: The methods have been described in Sect.

Siyuan Zhou 19/2/22 5:15 PM
已删除: 2.2.2... [23]

Siyuan Zhou 19/2/22 5:15 PM
已删除: ... [24]

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

974

975 **6.3 Uncertainties of different indicators**

976 In this study, we used two indicators to discriminate the different pollution levels, which are AOD and CDNC.
977 AOD is a good proxy for the large-scale pollution level, but it cannot well represent CCN (Shinozuka et al.,
978 2015). The value of AOD is influenced by moisture condition (Twohy et al., 2009). CDNC is a better proxy
979 for CCN, but it also has its uncertainties because it is calculated by the COT and CER. We can draw the same
980 conclusion on heavy rainfall diurnal changes between clean and polluted condition when using AOD and
981 CDNC respectively (Fig. 2). But when investigating the differences of cloud properties between clean and
982 polluted condition, there is a different result between using AOD and using CDNC, that the liquid CER is
983 decreased when CDNC increases (Fig. 6) while the liquid CER is increased when AOD increases. The
984 difference might be related with that the measurement biases, e.g., satellite AOD is evidently influenced by
985 the cloud (Brennan et al., 2005).

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

992

993 **7. Conclusions**

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

Siyuan Zhou 19/2/22 5:15 PM

已删除: found the starting and peak time of heavy rainfall occur earlier and the rainfall duration becomes longer under pollution.

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

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

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

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

.029

.030 Data availability

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

Siyuan Zhou 19/2/22 5:15 PM
已删除: and liquid CER ...re increa... [25]

Siyuan Zhou 19/2/22 5:15 PM
已删除: As a summary using a schematic diagram (Figure 10)...ccording to ill... [26]

Siyuan Zhou 19/2/22 5:15 PM
已删除: Although... This study h... [27]

Siyuan Zhou 19/2/22 5:15 PM
已删除: MERRA2 reanalysis...ltra... [28]

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

.098 | **Author contributions**

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

.102 | **Competing interests**

.103 | The authors declare that they have no conflict of interest.

.104 | **Acknowledgements**

.105 | This study is supported by funds from the National Key Research and Development Program-Global Change
.106 | and Mitigation Project: Global Change Risk of Population and Economic System: Mechanism and Assessment
.107 | (2016YFA0602401), the National Natural Science Foundation of China (grant nos. 41375003, 41621061 and
.108 | 41575143) and Project supported by State Key Laboratory of Earth Surface Processes and Resource Ecology
.109 | and Key Laboratory of Environmental Change and Natural Disaster. Wei-Chyung Wang acknowledges the
.110 | support of a grant (to SUNYA) from the Office of Sciences (BER), U.S. DOE.

.111 | **References:**

.112 | Albrecht, B. A.: Aerosols, cloud microphysics, and fractional cloudiness, *Science* 245: 1227-1230, 1989.
.113 | Anonymous: 1994. Atmospheric Sciences Thesaurus. China Meteorological Press: Beijing, China. (in
.114 | Chinese)
.115 | Anonymous (2013), IPCC fifth assessment report, *Weather*, 68, 310-310.
.116 | Bellouin, N., Quaas, J., Morcrette J. -J., and Boucher, O.: Estimates of aerosol radiative forcing from the
.117 | MACC re-analysis. *Atmos. Chem. Phys.*, 13: 2045-2062, 2013.
.118 | Benedetti, A., Morcrette, J. J., Boucher, O., Dethof, A., Engelen, R. J., Fisher, M., Flentje, H., Huneeus, N.,
.119 | Jones, L., Kaiser, J. W., Kinne, S., Mangold, A., Razinger, M., Simmons, A. J., and Suttie, M.: Aerosol
.120 | analysis and forecast in the European Centre for Medium-Range Weather Forecasts Integrated Forecast
.121 | System: 2. Data assimilation. *J. Geophys. Res.* 114: D13205 doi:10.1029/2008JD011115, 2009.
.122 | Brennan, J., Kaufman, Y., Koren, I., and Rong, L.: Aerosol-cloud interaction-Misclassification of MODIS
.123 | clouds in heavy aerosol, *IEEE T. Geosci. Remote*, 43, 911-915,
.124 | <https://doi.org/10.1109/TGRS.2005.844662>, 2005.
.125 | Bennartz, R., and Rausch, J.: Global and regional estimates of warm cloud droplet number concentration
.126 | based on 13 years of AQUA-MODIS observations, *Atmos. Chem. Phys.*, 17: 9815-9836, 2017.
.127 | Bennartz, R.: Global assessment of marine boundary layer cloud droplet number concentration from satellite, *J.*
.128 | *Geophys. Res.*, 112, D02201, doi:10.1029/2006JD007547, 2007.
.129 |

Siyuan Zhou 19/2/22 5:15 PM
已删除: disc

Siyuan Zhou 19/2/22 5:15 PM
已删除: daac-bin/FTPSubset2.pl

Siyuan Zhou 19/2/22 5:15 PM
已删除: CZ,

Siyuan Zhou 19/2/22 5:15 PM
已删除: CZ

Siyuan Zhou 19/2/22 5:15 PM
已删除: WCW

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符, 空格 段前: 0 pt, 段后: 0 pt

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM
已删除: Lawrence, B. N.: Regional

Siyuan Zhou 19/2/22 5:15 PM
已设置格式

Siyuan Zhou 19/2/22 5:15 PM
已删除: Thomas, G. E., Arnold, C. P. G., Campmany, E., Carboni,

Siyuan Zhou 19/2/22 5:15 PM
已下移 [5]: E.,

Siyuan Zhou 19/2/22 5:15 PM
已删除: Grainger, R. G., Poulsen,

Siyuan Zhou 19/2/22 5:15 PM
已下移 [6]: C.,

Siyuan Zhou 19/2/22 5:15 PM
已删除: Bulgin, C. E., Palmer, P.

Siyuan Zhou 19/2/22 5:15 PM
已删除: Siddans, R.,

Siyuan Zhou 19/2/22 5:15 PM
已设置格式

Siyuan Zhou 19/2/22 5:15 PM
已删除: seasonal variations of the Twomey indirect effect as observed by the ATSR-2

Siyuan Zhou 19/2/22 5:15 PM
已设置格式

Siyuan Zhou 19/2/22 5:15 PM
已删除: instrument,

Siyuan Zhou 19/2/22 5:15 PM
已设置格式

Siyuan Zhou 19/2/22 5:15 PM
已删除: . Lett. 35, L02811

Siyuan Zhou 19/2/22 5:15 PM
已设置格式

Siyuan Zhou 19/2/22 5:15 PM
已删除: 2007GL031394, 2008

.148 [Boers, R., Acarreta, J. A., and Gras, J. L.: Satellite monitoring of the first indirect aerosol effect: Retrieval of](#)
.149 [the droplet concentration of water clouds. *J. Geophys. Res.*, 111, D22208, doi:10.1029/2005JD006838,](#)
.150 [2006.](#)

.151 Chen, Q., Yin, Y., Jin, L., Xiao, H., and Zhu, S.: The effect of aerosol layers on convective cloud
.152 microphysics and precipitation, *Atmos. Res.*, **101**, 327-340, 2011.

.153 Cheng, C. T., Wang, W. C., and Chen, J. P.: A modeling study of aerosol impacts on cloud microphysics and
.154 radiative properties, *Q. J. R. Meteorol. Soc.*, 133, 283–297, doi:10.1002/qj.25, 2007.

.155 Cheng, C. T., Wang, W. C., and Chen, J. P.: Simulation of the effects of increasing cloud condensation nuclei
.156 on mixed-phase clouds and precipitation of a front system. *Atmos. Res.*, 96: 461-476, doi:
.157 10.1016/j.atmosres.2010.02.005, 2010.

.158 Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M.
.159 A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N.,
.160 Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E.
.161 V., Isaksen, I., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M.,
.162 Morcrette, J.-J., Park, B.-K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J.-N., Vitart, F.: The
.163 ERA-Interim reanalysis: configuration and performance of the data assimilation system. *Q. J. R.*
.164 *Meteorol. Soc.* 137: 553–597. doi:10.1002/qj.828, 2011.

.165 Fan, J. W., Rosenfeld, D., Yang, Y., Zhao, C., Leung, L. R., and Li, Z. Q.: Substantial contribution of
.166 anthropogenic air pollution to catastrophic floods in Southwest China. *Geophys. Res. Lett.* 42:
.167 6066-6075, 2015.

.168 Garrett, T. J. and Zhao, C.: Increased Arctic cloud longwave emissivity associated with pollution from
.169 mid-latitudes. *Nature* 440(7085): 787-9, 2006.

.170 Givati, A., and Rosenfeld, D.: Quantifying precipitation suppression due to air pollution. *J. Appl. Meteor.* 43:
.171 1038-1056, 2004.

.172 [Gryspeerd, E., Stier, P., and Partridge, D. G.: Links between satellite-retrieved aerosol and precipitation.](#)
.173 [Atmos. Chem. Phys., 14, 9677–9694, 2014.](#)

.174 Gunthe, S. S., Rose, D., Su, H., Garland, R. M., Achtert, P., Nowak, A., Wiedensohler, A., Kuwata, M.,
.175 Takegawa, N., Kondo, Y., Hu, M., Shao, M., Zhu, T., Andreae, M. O., and Poschl, U.: Cloud
.176 condensation nuclei (CCN) from fresh and aged air pollution in the megacity region of Beijing, *Atmos.*
.177 *Chem. Phys.* 11(21): 11023-11039, 2011.

.178 Guo, C. W., Xiao, H., Yang, H. L., and Tang, Q.: Observation and modeling analyses of the macro-and
.179 microphysical characteristics of a heavy rain storm in Beijing, *Atmos. Res.*, 156: 125-141, doi:
.180 10.1016/j.atmosres.2015.01.007, 2015.

.181 Guo, J. P., Deng, M. J., Lee, S. S., Wang, F., Li, Z. Q., Zhai, P. M., Liu, H., Lv, W., Yao, W., and Li, X. W.:
.182 Delaying precipitation and lightning by air pollution over the Pearl River Delta. Part I: Observational
.183 analyses. *J. Geophys. Res. Atmos.* 121: 6472-6488, 2016.

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 左, 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符, 孤行控制, 不调整西文与中文之间的空格, 不调整中文和数字之间的空格

Siyuan Zhou 19/2/22 5:15 PM
已设置格式

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM
已删除: DOI

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM
已删除: DOI

.186 | Guo, L., Highwood, E. J., Shaffrey, L. C., and Turner, A. G.: The effect of regional changes in anthropogenic
.187 | aerosols on rainfall of the East Asian Summer Monsoon. *Atmos. Chem. Phys.* 13: 1521-1534, 2013.

.188 | Guo, X. L., Fu, D. H., Guo, X., and Zhang, C. M.: A case study of aerosol impacts on summer convective
.189 | clouds and precipitation over northern China. *Atmos. Res.* 142: 142-157, 2014.

.190 | [Hammer, M. S., Martin, R. V., Li, C., Torres, O., Manning, M., and Boys, B. L.: Insight into global trends in
.191 | aerosol composition from 2005 to 2015 inferred from the OMI Ultraviolet Aerosol Index, *Atmos. Chem.
.192 | Phys.*, 18: 8097-8112, 2018.](#)

.193 | Harikishan, G., Padmakumari, B., Maheskumar, R. S., Pandithurai, G., and Min, Q. L.: Aerosol indirect effects
.194 | from ground-based retrievals over the rain shadow region in Indian subcontinent, *J. Geophys. Res. Atmos.*
.195 | 121(5): 2369-2382, 2016.

.196 | [Higgins, R. W., Yao, Y., Yarosh, E. S., Janowiak, J. E. and Mo, K. C.: Influence of the Great Plains low-level
.197 | jet on summertime precipitation and moisture transport over the central United States, *J. Climate*, 10,
.198 | 481-507, 1997.](#)

.199 | [Holz, R. E., Ackerman, S. A., Nagle, F. W., Frey, R., Dutcher, S., Kuehn, R. E., Vaughan, M. A., and Baum,
.200 | B.: Global Moderate Resolution Imaging Spectroradiometer \(MODIS\) cloud detection and height
.201 | evaluation using CALIOP, *J. Geophys. Res. Atmos.*, 113: D00A19, doi: 10.1029/2008JD009837, 2008.](#)

.202 | Jacobson, M. Z.: Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols,
.203 | *Nature* 409: 695-697, 2001.

.204 | Jiang, H., Feingold, G., and Cotton, W. R.: Simulations of aerosol-cloud-dynamical feedbacks resulting from
.205 | entrainment of aerosol into the marine boundary layer during the Atlantic Stratocumulus Transition
.206 | Experiment, *J. Geophys. Res.*, 107(D24), 4813, doi:10.1029/2001JD001502, 2002.

.207 | Jiang, J. H., Su, H., Schoeberl, M. R., Massie, S. T., Colarco, P., Platnick, S., and Livesey, N. J.: Clean and
.208 | polluted clouds: Relationships among pollution, ice clouds, and precipitation in South America, *Geophys.
.209 | Res. Lett.*, 35, L14804, doi: 10.1029/2008GL034631, 2008.

.210 | Jiang, M. J., Li, Z. Q., Wan, B. C., and Cribb, M.: Impact of aerosols on precipitation from deep convective
.211 | clouds in eastern China. *J. Geophys. Res.* 121: 9607-9620, 2016.

.212 | Johnson, D. B.: The role of giant and ultra-giant aerosol particles in warm rain initiation, *J. Atmos. Sci.*, 39,
.213 | 448-460, doi:10.1175/1520-0469(1982)039<0448:TROGAU>2.0.CO;2, 1982.

.214 | Jung, W. S., Panicker, A. S., Lee, D. I., and Park, S. H.: Estimates of aerosol indirect effect from Terra
.215 | MODIS over Republic of Korea, *Advances in Meteorology*, 2013 (976813): 1-8,
.216 | <http://dx.doi.org/10.1155/2013/976813>, 2013.

.217 | [Kim, K., -M., Lau, K. M., Sud, Y. C., and Walker, G. K.: Influence of aerosol radiative forcings on the diurnal
.218 | and seasonal cycles of rainfall over West Africa and Eastern Atlantic Ocean using GCM simulation. *Clim.
.219 | Dyn.* 35\(1\):115-126, doi: 10.1007/s00382-010-0750-1, 2010.](#)

.220 | Lau, K. M., Kim, M. K., and Kim, K. M.: Asian summer monsoon anomalies induced by aerosol direct
.221 | forcing: the role of the Tibetan Plateau. *Clim. Dyn.*, 26: 855-864, 2006.

.222 | Lee, S. S., Donner, L. J., and Phillips, V. T. J.: Impacts of aerosol chemical composition on microphysics and
.223 | precipitation in deep convection. *Atmos. Res.*, 94, 220-237, 2009.

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM
已移动(插入) [5]

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM
已移动(插入) [6]

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

- .224 | Lee, S. S., Guo, J., and Li, Z.: Delaying precipitation by air pollution over the Pearl River Delta: 2. Model
.225 | simulation. *J. Geophys. Res. Atmos.*, 121: 11739-11760, 2016.
- .226 | Lelieveld, J. and Heintzenberg, J.: Sulfate cooling effect on climate through in-cloud oxidation of
.227 | anthropogenic SO₂. *Science* 258: 117-120, 1992.
- .228 | Levy, R. C., Mattoo, S., Munchak, L. A., Remer, L. A., Sayer, A. M., Patadia, F., and Hsu, N. C.: The
.229 | Collection 6 MODIS aerosol products over land and ocean, *Atmos. Meas. Tech.*, 6, 2989–3034,
.230 | <https://doi.org/10.5194/amt-6-2989-2013>, 2013.
- .231 | [Li, H., Cui, X., Zhang, W., and Qiao, L.: Observational and dynamic downscaling analysis of a heavy rainfall
.232 | event in Beijing, China during the 2008 Olympic Games. *Atmos. Sci. Lett.*, 17, 368-376, 2016.](#)
- .233 | [Li, Z., Niu, F., Fan, J., Liu, Y., Rosenfeld, D., and Ding, Y.: Long-term impacts of aerosols on the vertical
.234 | development of clouds and precipitation, *Nat. Geosci.*, 4, 888-894, 2011.](#)
- .235 | Lim, K. S. and Hong, S.: Investigation of aerosol indirect effects on simulated flash-flood heavy rainfall over
.236 | Korea, *Meteor. Atmos. Phys.*, **118**, 199-214, 2012.
- .237 | Liu, G., Shao, H., Coakley Jr. J. A., Curry, J. A., Haggerty, J. A., and Tschudi, M. A.: Retrieval of cloud
.238 | droplet size from visible and microwave radiometric measurements during INDOEX: Implication to
.239 | aerosols' indirect radioactive effect, *J. Geophys. Res.*, 108(D1), 4006, doi:10.1029/2001JD001395, 2003.
- .240 | [Liu, J., Wang, S., Zhang, W., and Wei, X.: Mechanism analysis of a strong convective weather in Hebei
.241 | Province. *Advances in Marine Science*, 30, 9-16, 2012.](#)
- .242 | [Menzel, W. P., Frey, R. A., Zhang, H., Wylie, D. P., Moeller, C. C., Holz, R. E., Maddux, B., Baum, B. A.,
.243 | Strabala, K. I., and Gumley, L. E.: MODIS global cloud-top pressure and amount estimation: Algorithm
.244 | description and results, *J. Appl. Meteorol. Clim.*, 47\(4\):1175-1198, doi: 10.1175/2007JAMC1705.1,
.245 | 2008.](#)
- .246 | [Min, Q., Joseph, E., Lin, Y., Min, L., Yin, B., Daum, P. H., Kleinman, L. I., Wang, J., and Lee, Y. -N.:
.247 | Comparison of MODIS cloud microphysical properties with in-situ measurements over the Southeast
.248 | Pacific, *Atmos. Chem. Phys.*, 12: 11261-11273, 2012.](#)
- .249 | Panicker, A. S., Pandithurai, G., and Dipu, S.: Aerosol indirect effect during successive contrasting monsoon
.250 | seasons over Indian subcontinent using MODIS data, *Atmospheric environment* 44(15): 1937-1943,
.251 | 2010.
- .252 | Platnick, S., Meyer, K., King, M. D., Wind, G., Amarasinghe, N., Marchant, B., Arnold, G. T., Zhang, Z.,
.253 | Hubanks, P. A., Holz, R. E., Yang, P., Ridgway, W. L., and Riedi, J.: The MODIS cloud optical and
.254 | microphysical products: Collection 6 updates and examples from Terra and Aqua. *IEEE Trans. Geosci.*
.255 | *Remote Sens.*, 55, 502-525, doi:10.1109/TGRS.2016.2610522, 2017
- .256 | [Qian, Y., Gong, D. Y., Fan, J. W., Leung, L. R., Bennartz, R., Chen, D. L., Wang, W. G.: Heavy pollution
.257 | suppresses light rain in China: Observations and modeling, *J. Geophys. Res. Atmos.* **114**: D00K02, 2009.](#)
- .258 | Qiu, Y., Zhao, C., Guo, J., and Li, J.: 8-Year ground-based observational analysis about the seasonal variation
.259 | of the aerosol-cloud droplet effective radius relationship at SGP site. *Atmos. Environ.* 164: 139-146,

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

260 2017.

261 Rienecker, M. M., Suarez, M. J., Todling, R., Bacmeister, J., Takacs, L., Liu, H. C., Gu, W., Sienkiewicz, M.,

262 Koster, R. D., Gelaro, R., Stajner, I., Nielsen, J. E.: The GEOS-5 Data Assimilation

263 System—Documentation of Versions 5.0.1 and 5.1.0, and 5.2.0. NASA Technical Report Series on

264 Global Modeling and Data Assimilation NASA/TM-2008-104606 27: 92 pp, 2008.

265 Rosenfeld, D.: TRMM observed first direct evidence of smoke from forest fires inhibiting rainfall, *Geophys.*

266 *Res. Lett.*, 26, 3105–3108, doi:10.1029/1999GL006066, 1999.

267 Rosenfeld, D., Lohmann, U., Raga, G. B., O'Dowd, C. D., Kulmala, M., Fuzzi, S., Reissell, A., Andreae, M.

268 O.: Flood or drought: How do aerosols affect precipitation? *Science*, 321:1309-1313, 2008.

269 Rosenfeld, D., Sherwood, S., Wood, R., and Donner, L.: Climate effects of aerosol-cloud interactions. *Science*,

270 343: 379-380, 2014.

271 Rosenfeld, D., and Woodley, W. L.: Convective clouds with sustained highly supercooled liquid water down

272 to -37°C , *Nature*, 405, 440–442, doi:10.1038/35013030, 2000.

273 Sassen, K., Starr, D., Mace, G. G., Poellot, M. R., Melfi, S. H., Eberhard, W.L., Spinhirne, J. D., Eloranta, E.

274 W., Hagan, D. E., and Hallett, J.: The 5–6 December 1991 FIRE IFO II jet stream cirrus case study:

275 Possible influences of volcanic aerosols, *J. Atmos. Sci.*, 52, 97–123, doi:10.1175/1520-0469(1995)

276 052<0097:TDFIJI>2.0.CO;2, 1995.

277 Shen, Y., Xiong, A., Wang, Y., and Xie, P.: Performance of high-resolution satellite precipitation products

278 over China, *J. Geophys. Res.*, 115, D02114, doi:10.1029/2009JD012097, 2010.

279 Sherwood, S.: Aerosols and ice particle size in tropical cumulonimbus, *J. Clim.*, 15, 1051–1063,

280 doi:10.1175/1520-0442(2002)015<1051:AAIPSI>2.0.CO;2, 2002.

281 [Shinozuka, Y., Clarke, A. D., Nenes, A., Jefferson, A., Wood, R., McNaughton, C. S., Ström, J., Tunved, P.,](#)

282 [Redemann, J., Thornhill, K. L., Moore, R. H., Latham, T. L., Lin, J. J., and Yoon, Y. J.: The relationship](#)

283 [between cloud condensation nuclei \(CCN\) concentration and light extinction of dried particles:](#)

284 [indications of underlying aerosol processes and implications for satellite-based CCN estimates, *Atmos.*](#)

285 [Chem. Phys.](#), 15, 7585-7604, 10.5194/acp-15-7585-2015, 2015.

286 Song, X. L. and Zhang, G. J.: Microphysics parameterization for connective clouds in a global climate model:

287 Description and single-column model tests, *J. Geophys. Res. Atmos.*, 116, D02201, 2011.

288 Squires, P.: The growth of cloud drops by condensation: I. general characteristics, *Aust. J. Sci. Res., Ser. A*, 5,

289 66–86, 1952.

290 Squires, P., and Twomey, S.: A comparison of cloud nucleus measurements over central North America and

291 Caribbean Sea, *J. Atmos. Sci.*, 23, 401–404, doi: 10.1175/1520-0469(1966)023<0401:ACOCNM>

292 <2.0.CO;2, 1966.

293 [Sun, Y. L., Wang, Z. F., Du, W., Zhang, Q., Wang, Q. Q., Fu, P. Q., Pan, X. L., Li, J., Jayne, J., and Worsnop,](#)

294 [D. R.: Long-term real-time measurements of aerosol particle composition in Beijing, China: seasonal](#)

295 [variations, meteorological effects, and source analysis, *Atmos. Chem. Phys.*](#), 15: 10149-10165, 2015.

Siyuan Zhou 19/2/22 5:15 PM

已删除: ?.

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符, 孤行控制, 不调整西文与中文之间的空格, 不调整中文和数字之间的空格

Siyuan Zhou 19/2/22 5:15 PM

已设置格式

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2 字符, 首行缩进: -2 字符

- .297 [Tariq, S., and Ali, M.: Spatio-temporal distribution of absorbing aerosols over Pakistan retrieved from OMI on](#)
.298 [board Aura Satellite. Atmos. Pollution Res. doi: 10.5094/APR.2015.030, 2015.](#)
- .299 Tao, M. H., Chen, L. F., Wang, Z. F., Tao, J. H., Che, H. Z., Wang, X. H., and Wang, Y.: Comparison and
.300 evaluation of the MODIS Collection 6 aerosol data in China. J. Geophys. Res. Atmos. 120:6992-7005,
.301 2015.
- .302 Tao, W. K., Chen, J. P., Li, Z., Wang, C., and Zhang C.: Impact of aerosols on convective clouds and
.303 precipitation. Rev. Geophys., 50, RG2001/2012: 1-62, doi: 10.1029/2011RG000369, 2012.
- .304 [Torres, O., Bhartia, P.K., Herman, J.R., Ahmad, Z., Gleason, J.: Derivation of aerosol properties from satellite](#)
.305 [measurements of backscattered ultraviolet radiation: Theoretical basis. J. Geophys. Res. Atmos. 103:](#)
.306 [17099–17110, 1998.](#)
- .307 [Twohy, C. H., Coakley, J. A., and Tahnk, W. R.: Effect of changes in relative humidity on aerosol scattering](#)
.308 [near clouds. Journal of Geophysical Research: Atmospheres, 114, n/a-n/a, 10.1029/2008JD010991, 2009.](#)
- .309 Twomey, S.: The influence of pollution on the shortwave albedo of clouds, J. Atmos. Sci., 34, 1149–1152,
.310 doi:10.1175/1520-0469(1977)034<1149:TIOPTO>2.0.CO;2, 1977.
- .311 Wang, J., Feng, J., Wu, Q., and Z. Yan, Z.: Impact of anthropogenic aerosols on summer precipitation in the
.312 Beijing-Tianjin-Hebei urban agglomeration in China: Regional climate modeling using WRF-Chem. Adv.
.313 Atmos. Sci., **33**, 753-766, 2016.
- .314 Wang, Z., Guo, P., and Zhang, H.: A Numerical Study of Direct Radiative Forcing Due to Black Carbon and
.315 Its Effects on the Summer Precipitation in China. Climatic and Environmental Research, **14**, 161-171,
.316 2009.
- .317 [Wolyn, P. G., and Mckee, T. B.: The mountain plains circulation east of a 2-km-high north south barrier. Mon.](#)
.318 [Weather Rev., 122, 1490-1508, 1994.](#)
- .319 Wu, P., Ding, Y. H., and Liu, Y. J.: Atmospheric circulation and dynamic mechanism for persistent haze
.320 events in the Beijing-Tianjin-Hebei region, Adv. Atmos. Sci., 34(4): 429-440, 2017.
- .321 Yang, X., Zhao, C., Zhou, L., Li, Z., Cribb, M., and Yang, S.: Wintertime cooling and a potential connection
.322 with transported aerosols in Hong Kong during recent decades. Atmos. Res. 211: 52-61, 2018.
- .323 Yu, R. C., Zhou, T. J., Xiong, A. Y., Zhu, Y. J., and Li, J. M.: Diurnal variations of summer precipitation over
.324 contiguous China. Geophys. Res. Lett. 34: L017041, 2007.
- .325 Yuan, T., Li, Z., Zhang, R., and Fan, J.: Increase of cloud droplet size with aerosol optical depth: An
.326 observation and modeling study. J. Geophys. Res. Atmos., 113: D04201, 2008.
- .327 Yuan, W. H., Yu, R. C., Chen, H. M., Li, J., and Zhang, M. H.: Subseasonal Characteristics of Diurnal
.328 Variation in Summer Monsoon Rainfall over Central Eastern China. J. Climate 23:6684-6695, 2010.
- .329 Zhou, S., Yang, J., Wang, W. C., Gong, D., Shi, P., and Gao, M.: Shift of daily rainfall peaks over the
.330 Beijing– Tianjin– Hebei region: An indication of pollutant effects? Int. J. Climatol. 2018:1–10.
.331 <https://doi.org/10.1002/joc.5700>, 2018.
- .332 [Zeng, S., Riedi, J., Trepte, C. R., Winker, D. M., and Hu, Y. -X.: Study of global cloud droplet number](#)
.333 [concentration with A-Train satellites. Atmos. Chem. Phys., 14: 7125-7134, doi:](#)

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM

已删除: DOI

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM

已设置格式: 缩进: 左: 0 cm, 悬挂缩进: 2
字符, 首行缩进: -2 字符

Siyuan Zhou 19/2/22 5:15 PM

已删除: .

... [29]

Unknown

已设置格式: 字体: Times, 11 pt, 加粗

.337
.338
.339
.340
.341
.342
.343
.344
.345
.346
.347
.348
.349
.350
.351
.352
.353
.354
.355
.356
.357
.358
.359
.360
.361
.362
.363
.364
.365
.366
.367
.368
.369
.370
.371
.372

[10.5194/acp-14-7125-2014, 2014.](#)
[Zhu, Y., Rosenfeld, D., and Li, Z.: Under what conditions can we trust retrieved cloud drop concentrations in broken marine stratocumulus? J. Geophys. Res. Atmos., 123: 8754-8767, 2018.](#)

.373
.374
.375

.376
.377
.378
.379
.380
.381
.382

.383
.384
.385
.386
.387
.388

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	699.20	2544.87
AAI	OMI	2005-2012	0.13	0.52
SAI	OMI	2005-2012	- 0.13	- 0.35
BC AOD	MACC	2003-2012	0.04	0.06
Sulfate AOD	MACC	2003-2012	0.46	0.87
Absolute humidity at 850 hPa (g/m ³)	ERA-interim	2002-2012	8.97	12.19

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	24.5	23.5	22.4	- 0.7	- 2.1	P<0.05	P<0.05
Peak time (LST)	23.0	23.1	22.0	18.9	- 1.0	- 4.2	P<0.05	P<0.05
Duration (hours)	4.0	4.9	4.8	7.3	+ 0.8	+ 2.4	P<0.05	P<0.05
Intensity (0.1mm/hour)	164.9	167.0	169.6	162.0	+ 4.7	- 5.0	P>0.1	P>0.1

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 clean condition and polluted condition using two indicators of AOD and CDNC, and the differences and significances of differences 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%.

Unknown
已设置格式: 字体:Times, 11 pt, 加粗

Unknown
已设置格式: 字体:Times, 11 pt

Siyuan Zhou 19/2/22 5:15 PM
已删除: (b)
Siyuan Zhou 19/2/22 5:15 PM
已设置格式
Siyuan Zhou 19/2/22 5:15 PM
已设置格式
Siyuan Zhou 19/2/22 5:15 PM
已删除: (c) duration (units: hours) and (d)

.391
.392

.393
.394
.395
.396
.397
.398
.399
.400
.401
.402
.403
.404
.405
.406
.407
.408
.409
.410
.411
.412
.413
.414
.415
.416
.417
.418

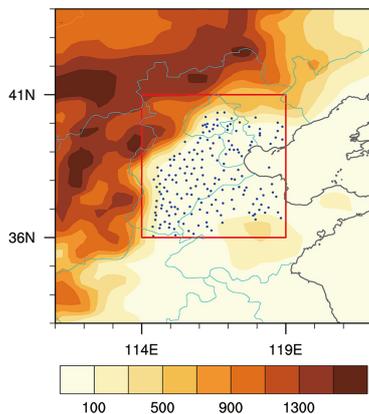
Group (case number)	CF	CTP	COT (liquid)	COT (ice)	CWP (liquid)	CWP (ice)	CER (liquid)	CER (ice)
1.Clean, dry (140)	67.9	460.3	4.9	4.1	62.5	80.4	21.1	34.6
2.Polluted, dry (75)	73.1 <small>0.05<p_{1,2}<0.1</small>	541.1	21.3	25.4	154.3	432.4	11.6	29.2
3.Clean, wet (191)	85.8	405.1	6.3	7.4	79.3	150.8	20.3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

Table 3. The average values of CF (units: %), CTP (units: hPa), COT (liquid and ice, units: none), CWP (liquid and ice, units: g/m²) and CER (liquid and ice, units: μm) in four groups. Grey numbers represent 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%.

Unknown
已设置格式: 字体:Times, 11 pt

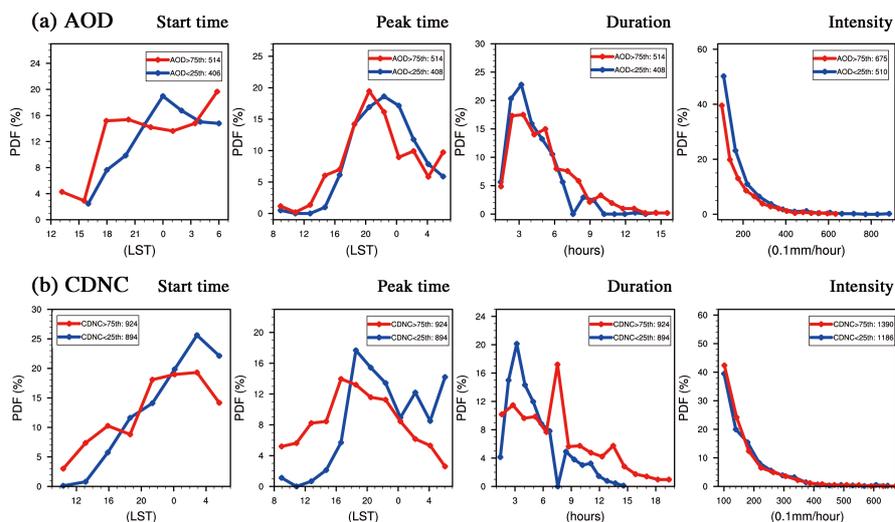
.419
.420
.421

Figures



.422
.423
.424
.425
.426

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



.427
.428
.429

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

Unknown

已设置格式: 字体:Times, 11 pt, 加粗

Unknown

已设置格式: 字体:Times, 11 pt, 加粗

Siyuan Zhou 19/2/22 5:15 PM

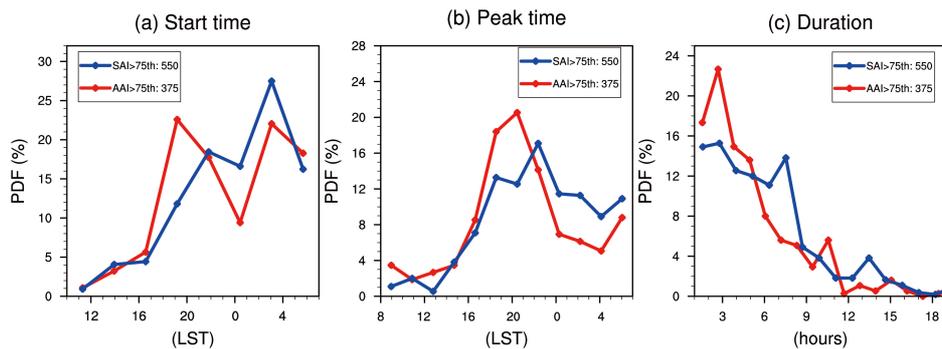
已删除: AOD<0.98

Siyuan Zhou 19/2/22 5:15 PM

已删除: :

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

.433



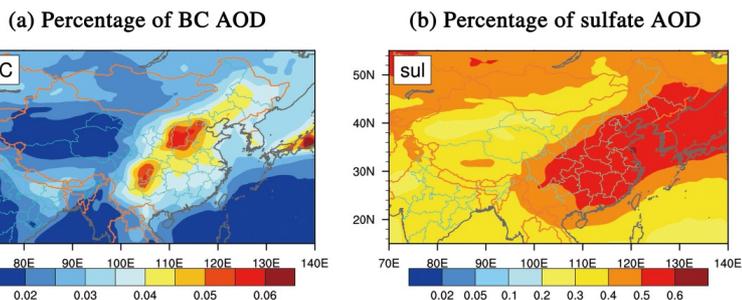
.434

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

.439

.440

.441



.442

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

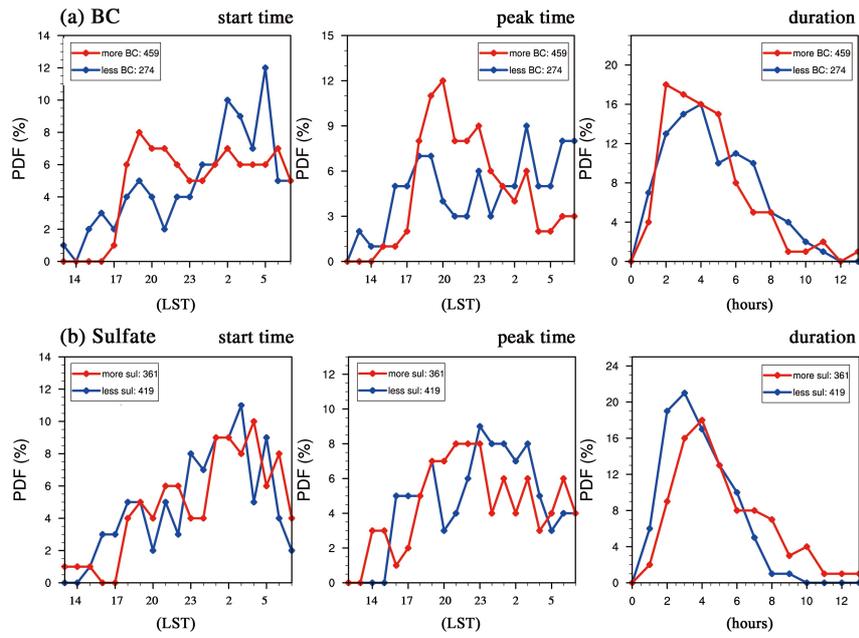
.444

.445

Siyuan Zhou 19/2/22 5:15 PM
已删除: >2.00) days, respectively,
Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 行距: 1.5 倍行距
Unknown
已设置格式: 字体: Times, 11 pt

Unknown
已设置格式: 字体: Times, 11 pt

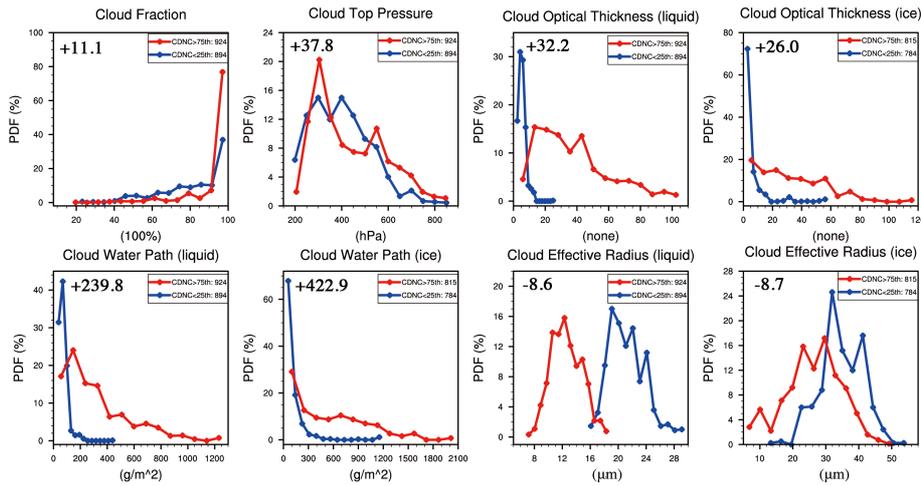
Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 定义网格后不调整右缩进,
行距: 1.5 倍行距, 不对齐到网格
Siyuan Zhou 19/2/22 5:15 PM
已移动(插入) [7]



Siyuan Zhou 19/2/22 5:15 PM
 已删除: . . . [30]
 Unknown
 已设置格式: 字体:Times, 11 pt
 Unknown
 已设置格式: 字体:Times, 11 pt

.447
 .448
 .449
 .450
 .451
 .452
 .453
 .454

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%.



.457

.458

.459

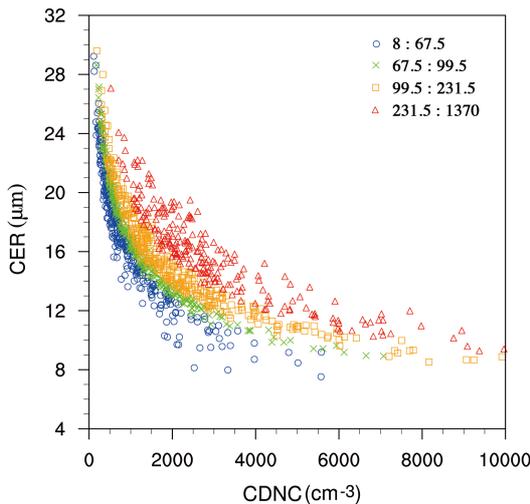
.460

.461

.462

.463

Figure 6. 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 lines: $CDNC < 25^{th}$ percentile) and polluted (red lines: $CDNC > 75^{th}$ percentile) heavy rainfall days. The numbers in the upper left stand for the mean differences between polluted and clean days (polluted minus clean). The differences between clean and polluted cases have all passed the significant test of 95%.



.464

.465

.466

Figure 7. Relationship of CER (units: μm) and CDNC (cm^{-3}) on different conditions of CWP (units: g/m^2). Different colors stand for different CWP conditions as shown in the legend.

Unknown
已设置格式: 字体:Times, 11 pt

Siyuan Zhou 19/2/22 5:15 PM
已删除: microns

Siyuan Zhou 19/2/22 5:15 PM
已删除: AOD<0.98

Siyuan Zhou 19/2/22 5:15 PM
已删除: AOD>2.00

Siyuan Zhou 19/2/22 5:15 PM
已删除: Here we removed the cases with the cloud top pressure more than 600hPa during 11 early summers (2002-2012).

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 定义网格后不调整右缩进, 行距: 1.5 倍行距, 不对齐到网格

Siyuan Zhou 19/2/22 5:15 PM
已删除: . . . [31]

Unknown
已设置格式: 字体:Times, 11 pt

Unknown
已设置格式: 字体:Times, 11 pt

Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 定义网格后不调整右缩进, 行距: 1.5 倍行距, 不对齐到网格

Siyuan Zhou 19/2/22 5:15 PM
已删除: 3. Diurnal variation

Siyuan Zhou 19/2/22 5:15 PM
已删除: cloud liquid water (units: mg/kg) respectively for (a) clean days, (b) polluted days and (c) difference (polluted minus clean), and cloud ice water (units: mg/kg) respectively for (d) clean days, (e) polluted days and (f) difference (polluted minus clean). Differences have passed 95% significant test. . . . [32]

Siyuan Zhou 19/2/22 5:15 PM
已删除: microns) in

Siyuan Zhou 19/2/22 5:15 PM
已删除:) on clean (blue lines: AOD<0.98) and polluted (red lines: AOD>2.00) days respectively for liquid and ice clouds

Unknown
已设置格式: 字体:Times, 11 pt

Unknown
已设置格式: 字体:Times, 11 pt

Unknown
已设置格式: 字体:Times, 11 pt

.488
.489

.490
.491
.492
.493
.494
.495
.496
.497
.498

.499
.500
.501
.502
.503

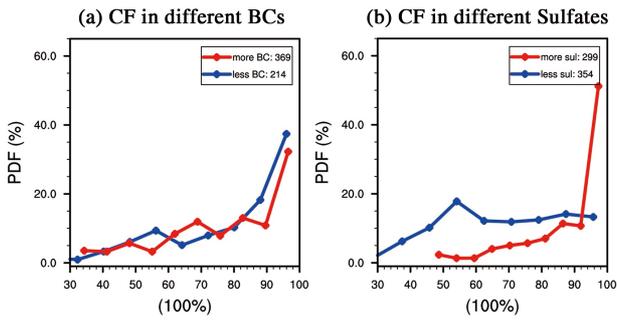


Figure 8.

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).

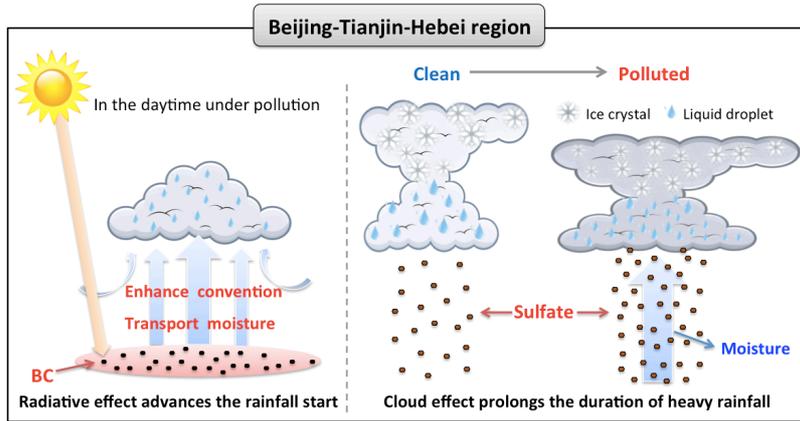


Figure 9. A schematic diagram for aerosols impact on heavy rainfall over Beijing-Tianjin-Hebei region.

已设置格式 ... [33]
Siyuan Zhou 19/2/22 5:15 PM
已删除: ... [34]
Unknown
已设置格式 ... [35]
Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 定义网格后不调整右缩进, 行距: 1.5 倍行距, 不对齐到网格
Siyuan Zhou 19/2/22 5:15 PM
已上移 [7]: . Percentages of AOD for (a) BC and (b) sulfate in JJA during 2002 to ... [36]
Siyuan Zhou 19/2/22 5:15 PM
(a) BC start tim
PDF (%) vs (LST) for more BC: 459 and less BC: 274
(b) Sulfate start tim
PDF (%) vs (LST) for more sul: 361 and less sul: 419
已删除: ... [37]
Unknown
已设置格式: 字体: Times, 11 pt
Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 行距: 1.5 倍行距
Unknown
已设置格式: 字体: Times, 11 pt
Siyuan Zhou 19/2/22 5:15 PM
已删除: and the cloud top pressure less than 600hPa
Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 两端对齐, 行距: 1.5 倍行距
Siyuan Zhou 19/2/22 5:15 PM
Enhance convection
Transport moisture
BC
Radiative effect advances the rain
已删除:
Unknown
已设置格式: 字体: Times, 11 pt
Unknown
已设置格式: 字体: Times, 11 pt
Siyuan Zhou 19/2/22 5:15 PM
... [38]
Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 行距: 1.5 倍行距, 不对齐到网格
Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 缩进: 首行缩进: 0 cm
Siyuan Zhou 19/2/22 5:15 PM
已设置格式: 定义网格后不调整右缩进, 行距: 1.5 倍行距, 不对齐到网格