Anonymous Referee #1

We greatly appreciate the constructive review from the referee that has improved the quality of our manuscript. We have considered each comment carefully and revised our manuscript accordingly to address the issues raised. Below we address each comment point by point. Reviewer comments are marked as black, our response as blue and changes to the manuscript as red.

This study investigated aerosol impacts on cloud and precipitation over northern Taiwan using aerosol and cloud datasets from Aqua/MODIS and surface measurements. The authors showed statistical analysis including the susceptibility of cloud droplet effective radius (CER) to aerosols (ACI), correlations between CER and cloud-top temperature, and size distributions of rain drop to find some signatures of aerosol-induced changes to cloud and precipitation properties. Although the analysis results shown tend to be consistent with one another and thus appear to suggest the aerosol impacts on cloud and precipitation over the target region, most of the analysis approach and the results shown, including the ACI analysis, relationships between rainfall and cloud water path, and CER-CTT joint statistics, are pretty much similar to what has already been done in a number of previous studies. I found no substantial novelty in materials included in the manuscript of its current form that deserves publication. Based on these evaluations, I cannot recommend the manuscript be considered for publication in Atmospheric Chemistry and Physics at least in its current form. One possible way for improving the overall study is to obtain a process-level insight into aerosol impacts on drizzle and precipitation exploiting the surface measurement of size distributions of rainfall, which might add some novelty to this study. Listed below are some specific points that (hopefully) might help the authors to re-construct their work in this direction for future potential submission of the revised manuscript.

We really appreciate and agree with these suggestions and comments from the referee. We have strengthened the analysis, in particular, the process-level insight into aerosol impacts on drizzle and precipitation by exploiting the surface measurement of rainfall size-distributions (lines: 257-283). As suggested, the analysis of ACI and CER-CTT statistics in terms of size-resolved characteristics of precipitation processes were included to support the discussion (lines: 206-215). We have addressed the specific comments in the sections below and made the revisions to the manuscript accordingly.

In addition, we believe our target region may be unique and stand out from other previous studies. First, the study area is located in the northwest Pacific Ocean where there has been much attention on aerosol transportation, as well as aerosol-cloud interactions from the literature (Tsay et al., 2016; Dong et al., 2019). However, observational-based studies are still lacking in this region. Second, this study integrates long-term satellite and surface measurements to assess ACI over a moderately polluted environment with complex terrain. Although the overall result appears similar to previous studies, it has important implications for the crucial role of cloud microphysics on the water cycle/resources in subtropical East Asia environment.

Specific comments:

- A novel piece of material included in the manuscript is rain drop size distribution measured by the JWD disdrometer, which should provide useful observation-based information for process-level assessment of the aerosol indirect effect on precipitation, i.e. how precipitation processes are modulated by aerosols. I would suggest the authors to conduct more detailed analysis of the rain drop size distributions and their relationships to differing conditions of aerosols, rather than just showing the simple plot of Fig. 10. Such an analysis should offer size-dependent view of aerosol impact on drizzle and precipitation and thus more in-depth insight into microphysics of the aerosol indirect effect.

Many thanks for this suggestion. We have added a more detailed analysis of the raindrop size distributions and the aerosol impact on drizzle and precipitation via the aerosol indirect effect. The paragraph has now been rewritten (lines: 257-283) and revised the original Fig. 10 to Fig. 10 and Fig. 11 as below:

Figure 10a shows the number of sample occurrences under different raindrop size classifications for clean and polluted days. The sample number (days) was significantly higher for clean conditions, suggesting rainfall was more common on clean days than on polluted days. We further calculated the minute-averaged droplet number in each raindrop size classification for polluted and clean days. Higher populations of raindrops were observed from bins n1 to n4, with the peak in bin n2 for both clean and polluted days (Fig. 10b). The difference is plotted in Fig. 10c. The results illustrate (Fig. 10c) that during polluted days, the droplet numbers appear lower for the smaller raindrop bins (≤ n8) compared to clean days and higher for the larger raindrop bins (> n8). A significant reduction in droplet number (decreased from 68 min^{-1} on clean days to 56 min^{-1} on polluted days) was observed in the n2 bin, corresponding to a reduction in drizzle. Our preliminary findings suggest that CCN may have competing effects (Ghan et al., 1998) on water uptake under aerosol-laden air and cloud water content-limited conditions, which would alter the precipitation processes.

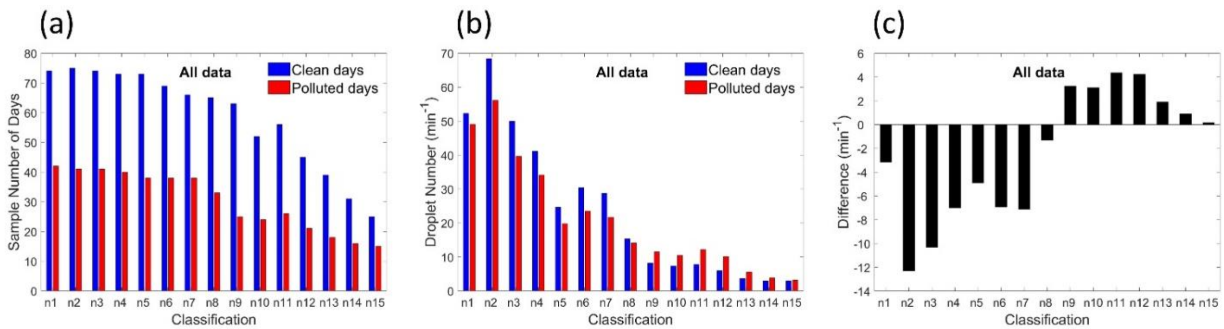

Figure 10: Multiyear (2005-2017) (a) JWD sample number of days in each raindrop size bin, (b) mean droplet number per minute for clean and polluted days and (c) The differences in the mean droplet number between polluted and clean days. nX reflects different raindrop size bins. The mean droplet size for n1 to n15 are, in order, 0.359, 0.455, 0.551, 0.656, 0.771, 0.913, 1.116, 1.331, 1.506, 1.665, 1.912, 2.259, 2.584, 2.869, and 3.198 mm.

To investigate the aerosol impacts on the change in droplet size, the cumulative number distribution of each raindrop size for clean and polluted days was calculated. We then normalized the data by computing the percentage of droplet numbers in each raindrop size class to the total number and the difference between polluted and clean days was defined by Eq. (2).

$$\text{nX Difference } (\%\, min^{-1}) = \frac{\sum_{i=1}^{dp} nX_i}{\sum_{X=1}^{b}\sum_{i=1}^{dp} nX_i} \times 100\ \% - \frac{\sum_{i=1}^{dc} nX_i}{\sum_{X=1}^{b}\sum_{i=1}^{dc} nX_i} \times 100\ \%, \tag{2}$$

where nX represents different raindrop size bins and b reflects the number of bins, b = 1-20; dp and dc represent the number of polluted and clean days respectively. The results are similar with Fig. 10c; the droplet numbers, on polluted days compared to clean days, appear lower for the smaller raindrop bins (≤ n5) and higher for the larger raindrop bins (> n5) (Fig. 11a). To investigate the aerosol impacts on light rain, we created a similar plot as Fig. 11a but only considered precipitation less than or equal to 1 mm h^{-1}, as shown in Fig. 11b. Our statistics for the droplet number concentration indicated that raindrop occurrence at n1 and n2 (i.e. drizzle) accounted for over 50 % on both polluted and clean days (not shown here) (shown as Fig. R1 in this response, but not shown in the revised manuscript), indicating that drizzle drops were a common raindrop type when rainfall was ≤ 1 mm h^{-1}. We determined that when rainfall was ≤ 1 mm h^{-1}, polluted days accounted for a more significant proportion of n1 and n2 than clean days (especially in the raindrop size distribution n1, which accounted for 2.3 %) (Fig. 11b). On the other hand, a decreased proportion of n3 to n8 was observed during polluted days, as compared with clean days. These results indicate that if precipitation is lower than or equal to 1 mm h^{-1} (i.e. light rain), abundant CCN drives raindrops to move towards smaller drop sizes, which increases the appearance of drizzle drops.

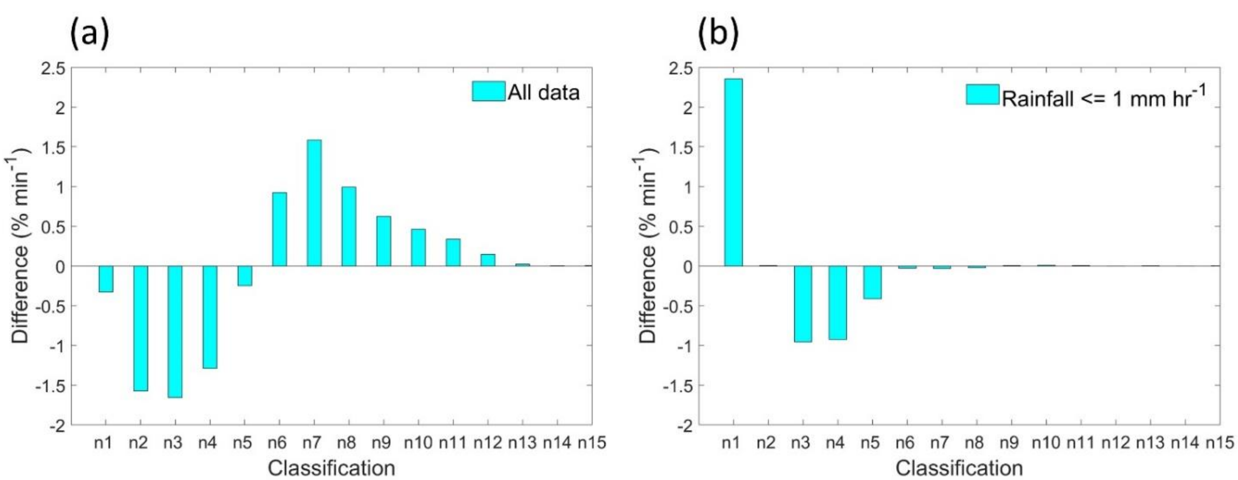

Figure 11: Multiyear (2005-2017) differences between polluted and clean days as percentages of the cumulative droplet number distribution for (a) all data and (b) the data with precipitation less than or equal to 1 mm h^{-1}. nX reflects different raindrop size bins as listed in Fig. 10.

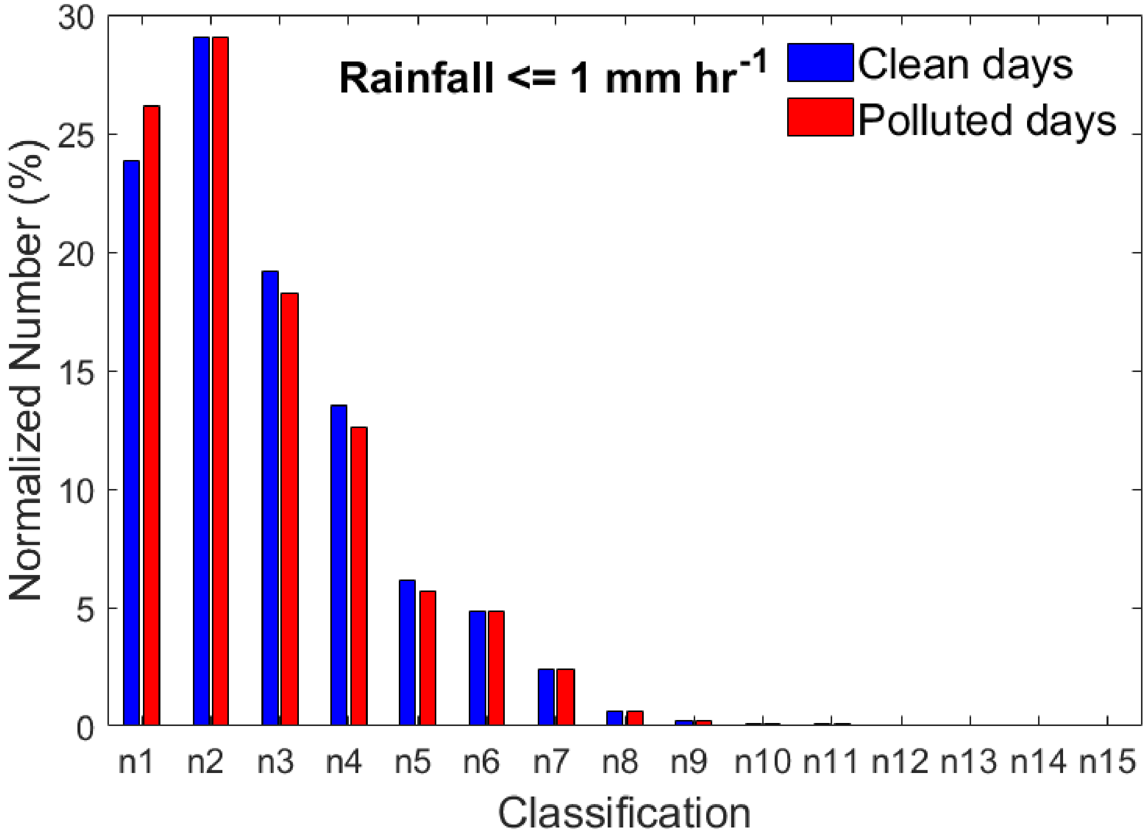

Figure R1: Multiyear (2005-2017) cumulative droplet number distribution for the JWD data for precipitation less than or equal to 1 mm h^{-1} on clean and polluted days. nX reflects different raindrop size bins as specified in Fig. 10.

- The size-resolved precipitation analysis might also add new insight into the analysis shown in Fig. 11. The statistics shown in Fig. 11a is quite similar to those already shown by satellite statistics of Lebsock et al. (2008) and L'Ecuyer et al. (2009), except that the authors' plot shows the rainfall rate (in ordinate) based on surface measurement, contrary to probability of precipitation in the two previous studies. I would suggest the statistics shown in Fig. 11a be broken down into different bins of drop size to see how the cloud-to-precipitation process varies with aerosols and how it depends on particle size of drizzle and rain. Such an analysis might offer a new process-level insight into the aerosol-induced suppression of precipitation. The same approach could also be applied to the analysis of Fig. 11b to obtain a "size-resolved view" of the temporal trend of precipitation and its relationship to aerosols.

Many thanks for this suggestion. We followed the suggestion and binned the rainfall data into drop size to study how the cloud-to-precipitation process varies with aerosol concentration and how it depends on the particle size of drizzle and rain. We divided the droplet bins into three groups: n1-n20, n1-n2, and n3-n20, representing all droplets, drizzle drops, and raindrops, respectively. We calculated the minute-averaged droplet number in each group of bins. The results shown in Fig. R2a, b, c demonstrate that the mean droplet number difference between polluted and clean days varies greatly between CWP groups 1–7, which may be due to the smaller sample number in each CWP group. However, whether drizzle drops or raindrops, the mean droplet number on clean days consistently exhibited higher values in CWP groups 8–10 compared with polluted days and increased with increasing CWP. In CWP group 9 ($150 \leq CWP < 297$), the mean droplet number on polluted days (12 min^{-1}) was lower by 38 min^{-1} compared with clean days (50 min^{-1}) when considering all droplets (Fig. R2a).

Figure R2d, e, f shows the 24-hour mean droplet number trends for CWP group 9 ($150 \leq CWP < 297$) on clean and polluted days, providing insights on the effect of aerosols on cloud lifetime. On clean days, when considering all droplets (n1-n20), the droplet number was larger than 50 min^{-1} except at 12:00, 20:00-23:00 and 02:00-03:00, whereas few droplets were observed during daytime on polluted days, and a droplet number greater than 50 min^{-1} registering only sporadically after 23:00. Considering raindrops (n3-n20), there was a notably larger droplet number observed after 03:00 (Fig. R2f). This may have been caused by high aerosol loading suppressing the precipitation in the daytime, delaying rainfall occurrence and in turn increasing the droplet number of larger raindrops in the early morning.

The above-mentioned results are in agreement with our revised manuscript discussing aerosol effects on precipitation (in Sect. 3.4), and suggesting precipitation might be suppressed and delayed under high aerosol loading. To avoid confusion for readers, this revised manuscript does not include the supplementary analysis described above.

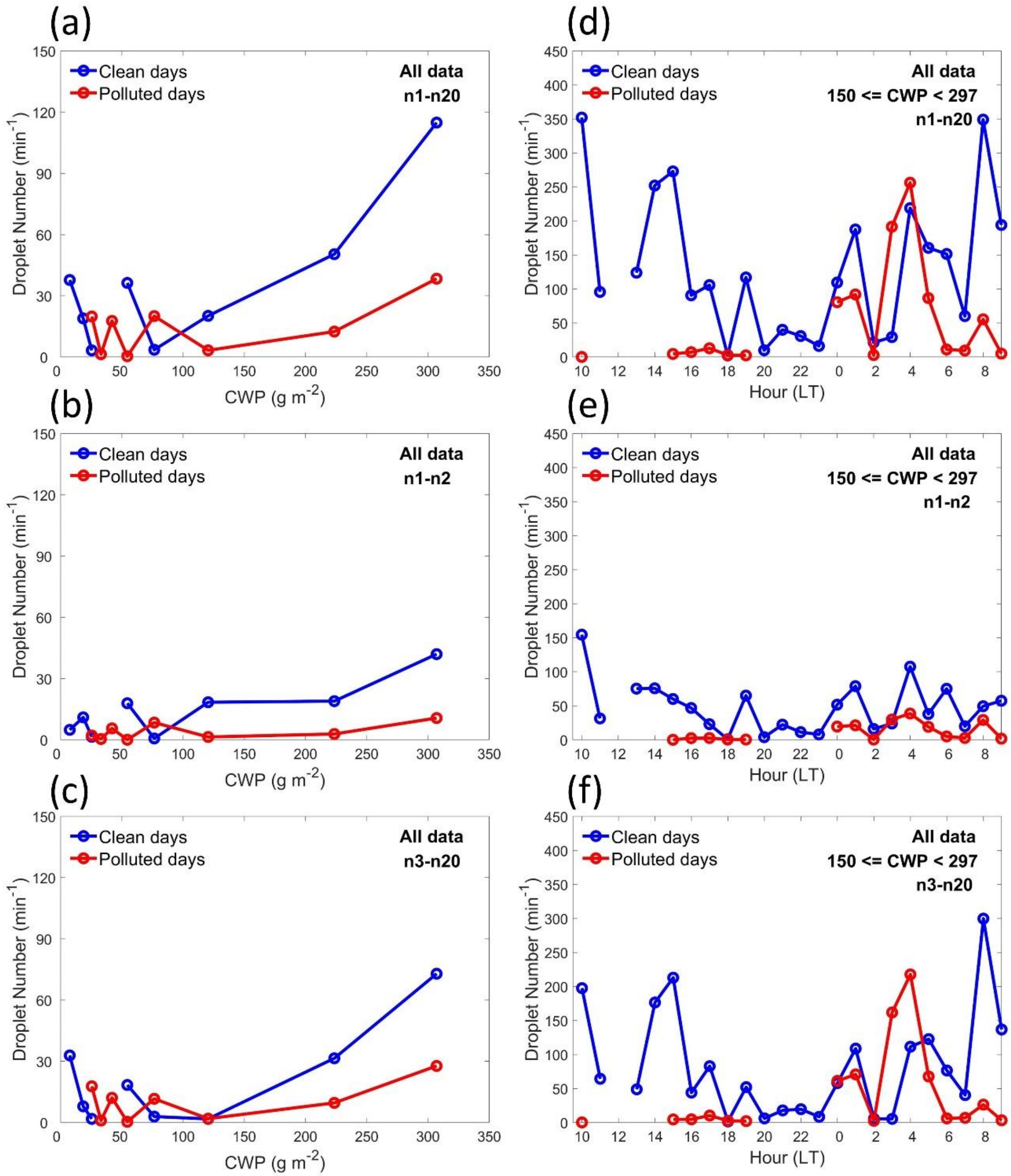

Figure R2: Multiyear (2005-2017) mean droplet number for (a) all droplets, (b) drizzle drops, and (c) raindrops in different CWP groups calculated for clean and polluted days. Hourly trend of mean droplet number for (d) all droplets, (e) drizzle drops, and (d) raindrops calculated for clean and polluted days when considering CWP group 9 ($150 \leq CWP < 297$) only.

- The joint statistics between CER and CTT shown in Fig. 8 are hard to interpret in its current form. I guess that the authors like to claim different CTT-CER correlations between clean and polluted conditions in Fig. 8a, but the tendency looks quite ambiguous in the plot shown. I would suggest apply analysis methodology of Rosenfeld and colleagues (e.g. Rosenfeld and Lensky, 1998; Rosenfeld 2000) that plot the mean and variance of CER at each CTT bin separately for clean and polluted conditions. It might show more clearly what the authors want to illustrate.

Thank you for the valuable suggestion. We now reference the analysis methodology of Rosenfeld (2000), and plot the mean and one standard deviation of CER at each CTT bin. The paragraph has been rephrased as below (lines: 206-215 in the revised manuscript):

The relationship between CTT and CER and aerosols was studied in further detail. Figure 8 displays CWP group 9 ($150 \leq CWP < 297$) results of the corresponding CTT-CER relationship and the occurrence frequency (%) of the CTT on clean and polluted days. On clean days, the mean CER increased from 10.7 to 12.7 μm as CTT decreased from 291 to 279 K, indicating an inverse relationship over much of the CTT range. This phenomenon could be caused by the onset of water cloud generation during strong updrafts, i.e. droplet size increases during air parcel expansion in an adiabatic process (Saito et al., 2019). However, on polluted days, as CTT lowered, the mean CER decreased; at CTT from 291 to 279 K, the CER decreased from 10.8 to 9.1 μm. Figure 8b shows that CTT exhibited a higher occurrence frequency between 288 and 285 K on polluted days, whereas clean days had a higher frequency of CTT between 285 and 282 K. These results suggest that abundant aerosols activated higher concentrations of CCN near the surface, which tends to form more low-level clouds with smaller cloud droplet size.

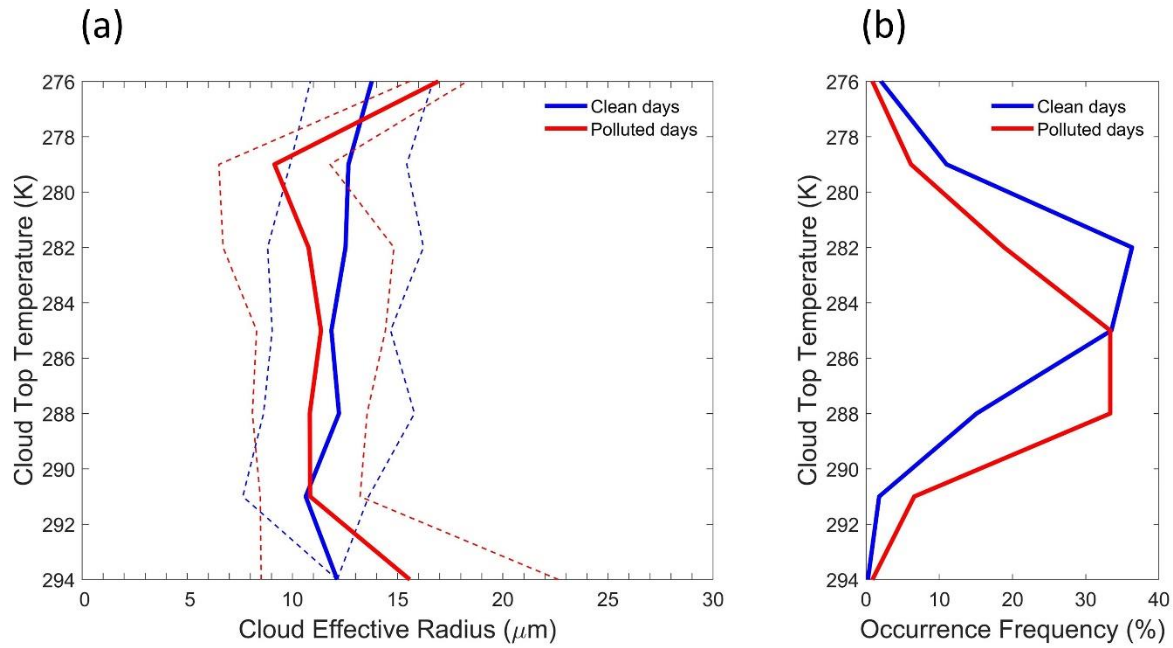

Figure 8: Multiyear (2005–2017) (a) cloud top temperature (CTT)-cloud effective radius (CER) relationship. Plotted are the mean (solid line) and one standard deviation (dashed line) of the CER for each 3 K interval, and (b) Frequency of occurrence of the CTT. Clean and polluted days are depicted with blue and red lines, respectively. Both (a) and (b) are constrained to CWP group 9 ($150 \leq CWP < 297$).

- These analyses proposed above could then be combined to enable interpreting the traditional analysis such as the ACI and CER-CTT statistics in terms of size-resolved characteristics of precipitation processes. Such an analysis would connect some of the existing metrics of the aerosol indirect effect in the context of precipitation processes, which would bring a valuable progress in understanding aerosol impacts on cloud and precipitation.

Thanks for the comments. Complementing the revisions mentioned above, the conclusions have been rephrased as (lines: 313-331 in the revised manuscript):

We used surface $PM_{2.5}$ mass concentration data as aerosol proxy to study the aerosol impacts on clouds and precipitation. According to $PM_{2.5}$ concentration level, the data was split into clean and polluted days. The analysis of aerosol effects on clouds indicated that in CWP group 9 (150 ≤

CWP < 297), the average COT in the main research area increased by 9.53, CER decreased by 2.77 μm, CF increased by 0.07, and CTT decreased by 1.28 K on polluted days compared with clean days. According to the aerosol indirect effect, polluted atmospheric conditions are connected with clouds characterized by lower CER, CTP, and larger CF and COT, which our results further support. Regarding the vertical distribution, our evidence shows that excess aerosols produced more liquid particles at lower altitude and inhibited the cloud droplet size under polluted conditions. Moreover, the effects of aerosol on cloud microphysics in polluted (i.e. land) and remote (i.e. ocean, less polluted) areas were investigated in CWP group 9, the ACI value of the remote area was 0.09, and the polluted area was 0.06. The ACI value in the remote area was larger than in the polluted area, indicating that clouds in the remote area were more sensitive to aerosol indirect effects.

Our analysis shows that precipitation might be suppressed and delayed under high aerosol loading. The observational data shows higher aerosol concentration redistributed cloud water to more numerous and smaller droplets under a constant liquid water content, reducing collision–coalescence rates, which further suppressed the precipitation and delayed rainfall duration. Our results are consistent with the cloud lifetime effect. Finally, we combined the observation of raindrop size distribution to complete the story of aerosol-cloud-precipitation interactions. As a result, on polluted days compared to clean days, droplet numbers decreased for smaller droplets bins but increased for larger droplets. However, when we looked into the light rain (≤ 1 mm h^{-1}) category, high concentration of aerosols drove raindrops towards smaller droplet sizes and increased the appearance of drizzle drops.

Minor points:

- Page 5, Line 27: COT should have no unit.

Thank you for correcting our mistakes. The sentence has been rephrased as (lines: 154-156):

The mean CWP, CF and CER in our study area ranged from 60–120 g m^{-2}, 0.6–0.7, and 13–14.5 μm, respectively. **COT was usually around 10** and most of the CTP was higher than 850 hPa, suggesting low-level clouds (e.g., warm, thin, and broken clouds).

- Page 6, Line 30: radiuses -> radii

Thank you for the correction. The sentence has been rephrased as (lines: 185-186):

The negative correlation for these groups indicates an aerosol indirect effect (i.e. an increase in aerosols cause cloud droplet **radii** to become smaller under a fixed water content).

- Page 7, Line 15: Does "cloud vertical profiles" mean CTT? It is not really the vertical profile but just a cloud-top temperature.

We agree with the reviewer's insight that CTT is not really the vertical profile but just a cloud top temperature. The paragraph has been rephrased as (lines: 206-215):

The relationship between CTT and CER and aerosols was studied in further detail. Figure 8 displays CWP group 9 ($150 \leq CWP < 297$) results of the corresponding CTT-CER relationship and the occurrence frequency (%) of the CTT on clean and polluted days. On clean days, the mean CER increased from 10.7 to 12.7 μm as CTT decreased from 291 to 279 K, indicating an inverse relationship over much of the CTT range. This phenomenon could be caused by the onset of water cloud generation during strong updrafts, i.e. droplet size increases during air parcel expansion in an adiabatic process (Saito et al., 2019). However, on polluted days, as CTT lowered, the mean CER decreased; at CTT from 291 to 279 K, the CER decreased from 10.8 to 9.1 μm. Figure 8b shows that CTT exhibited a higher occurrence frequency between 288 and 285 K on polluted days, whereas clean days had a higher frequency of CTT between 285 and 282 K. These results suggest that abundant aerosols activated higher concentrations of CCN near the surface, which tends to form more low-level clouds with smaller cloud droplet size.

- Figures 2, 5, 6, 9 and 11a: The horizontal axis for CWP should be logarithmic for at least some of the figures.

Thank you for the suggestion. We re-plotted the horizontal axis in logarithmic CWP and CWP group as shown below in Fig. R3. Although they have similar patterns, after our internal discussion, we decided to re-plot the original Fig. 6 and Fig. 9 with an x-axis of CWP group in the revised manuscript.

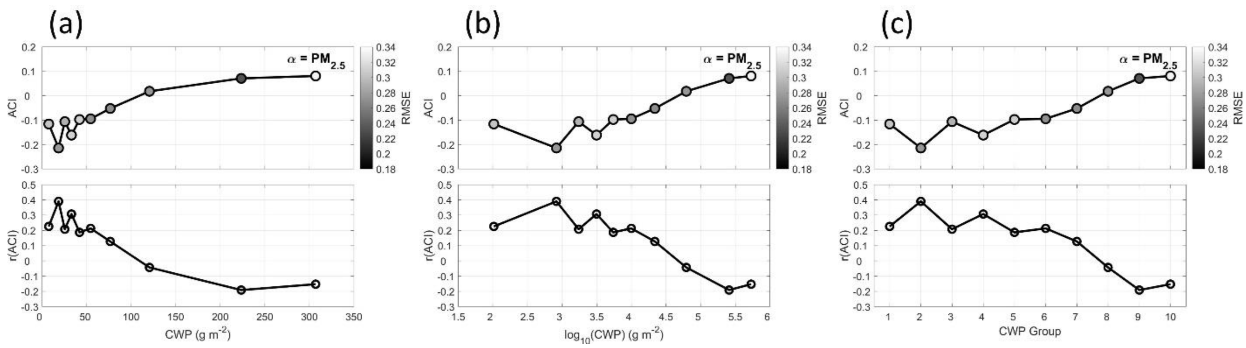

Figure R3: Aerosol cloud interaction estimated values vs. three different CWP variables (a) CWP, (b) logarithmic CWP, and (c) CWP group number.

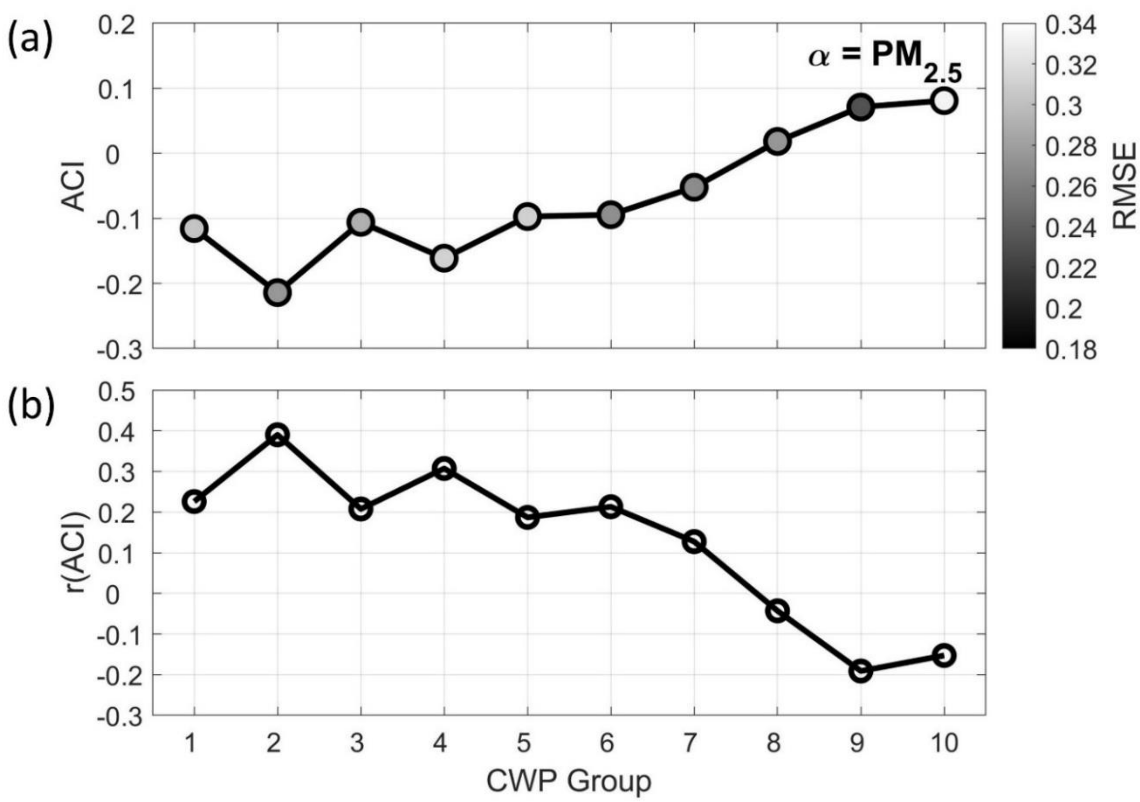

Figure 6: (a) Aerosol cloud interaction (ACI) estimated values, computed for the cloud effective radius (CER) in the different CWP groups by applying $PM_{2.5}$ concentrations as aerosol proxies. The shading in (a) represents the RMSE. (b) The correlation coefficients between $PM_{2.5}$ and CER are illustrated.

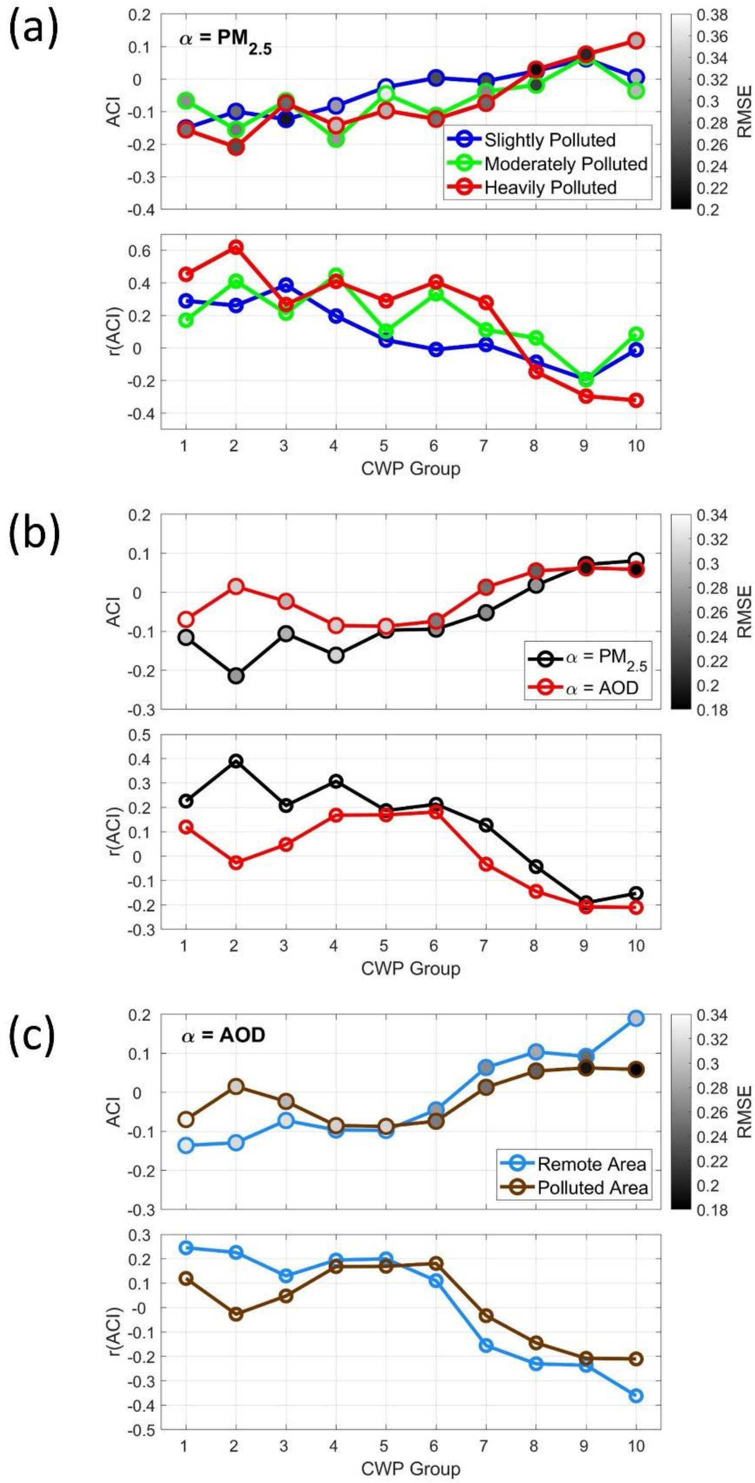

Figure 9: Multiyear (2005–2017) ACI values with the RMSE (shaded) and the correlation coefficient among (a) different polluted levels, (b) different aerosol proxies, and (c) different polluted condition areas.

Reference is cited in the response letter:

Dong, B., Wilcox, L. J., Highwood, E. J., and Sutton, R. T.: Impacts of recent decadal changes in Asian aerosols on the East Asian summer monsoon: roles of aerosol–radiation and aerosol–cloud interactions, Climate Dynamics, 53, 3235-3256, 2019.

Rosenfeld, D.: Suppression of rain and snow by urban and industrial air pollution, Science, 287, 1793-1796, 2000.

Tsay, S.-C., Maring, H. B., Lin, N.-H., Buntoung, S., Chantara, S., Chuang, H.-C., Gabriel, P. M., Goodloe, C. S., Holben, B. N., and Hsiao, T.-C.: Satellite-surface perspectives of air quality and aerosol-cloud effects on the environment: An overview of 7-SEAS/BASELInE, Aerosol and Air Quality Research, 16, 2581-2602, 2016.

New reference is added in the revised manuscript:

Ghan, S. J., Guzman, G., and Abdul-Razzak, H.: Competition between sea salt and sulfate particles as cloud condensation nuclei, Journal of the atmospheric sciences, 55, 3340-3347, 1998.