# Response to the reviewer 1 comments for "Aerosol impacts on warm-cloud microphysics and drizzle in a moderately polluted environment" by Chen et al.

**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.

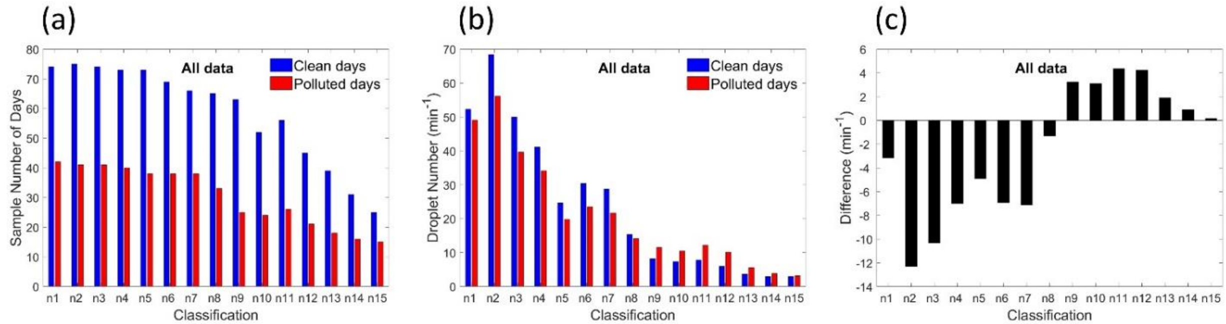



Figure 10: Multiyear (2005-2017) (a) JWD sample number of days in each raindrop size bin, (b) mean droplet number per minute for clean and polluted days and (c) The differences in the mean droplet number between polluted and clean days. nX reflects different raindrop size bins. The droplet size for n1 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).

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

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.

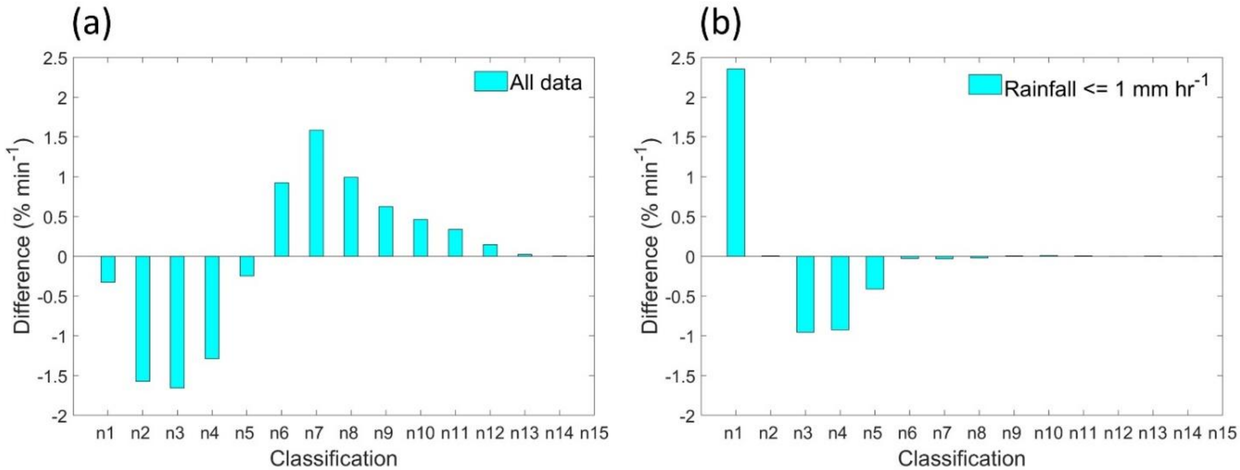



Figure 11: Multiyear (2005-2017) differences between polluted and clean days as percentages of the cumulative droplet number distribution for (a) all data and (b) the data with precipitation less than or equal to 1 mm $h^{-1}$. nX reflects different raindrop size bins as listed in Fig. 10.

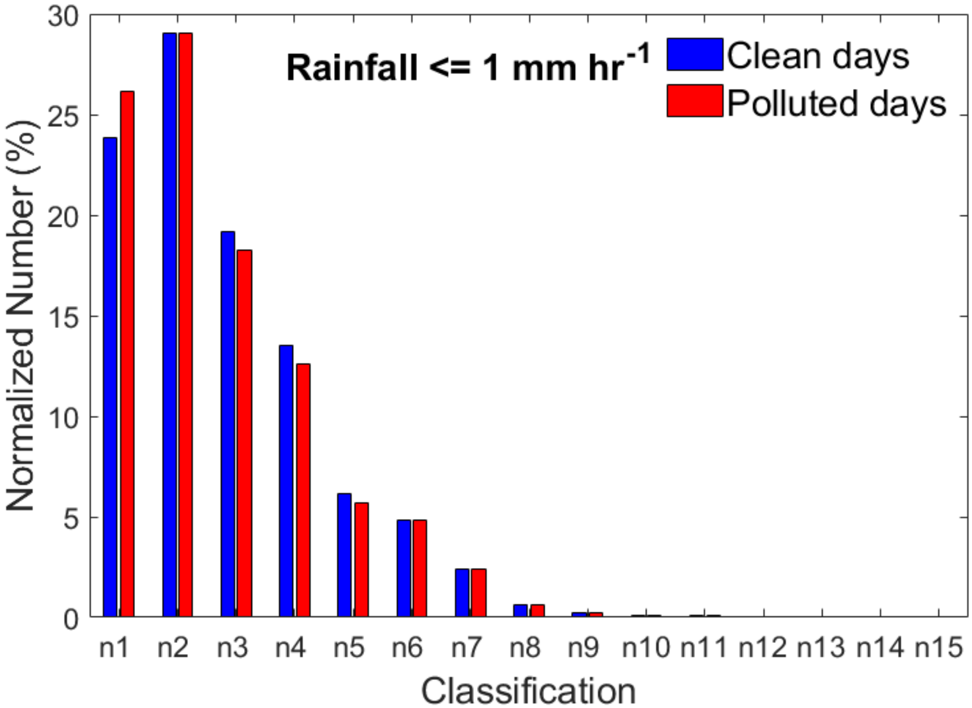



Figure R1: Multiyear (2005-2017) cumulative droplet number distribution for the JWD data for precipitation less than or equal to 1 mm $h^{-1}$ on clean and polluted days. nX reflects different raindrop size bins as specified in Fig. 10.

- 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.

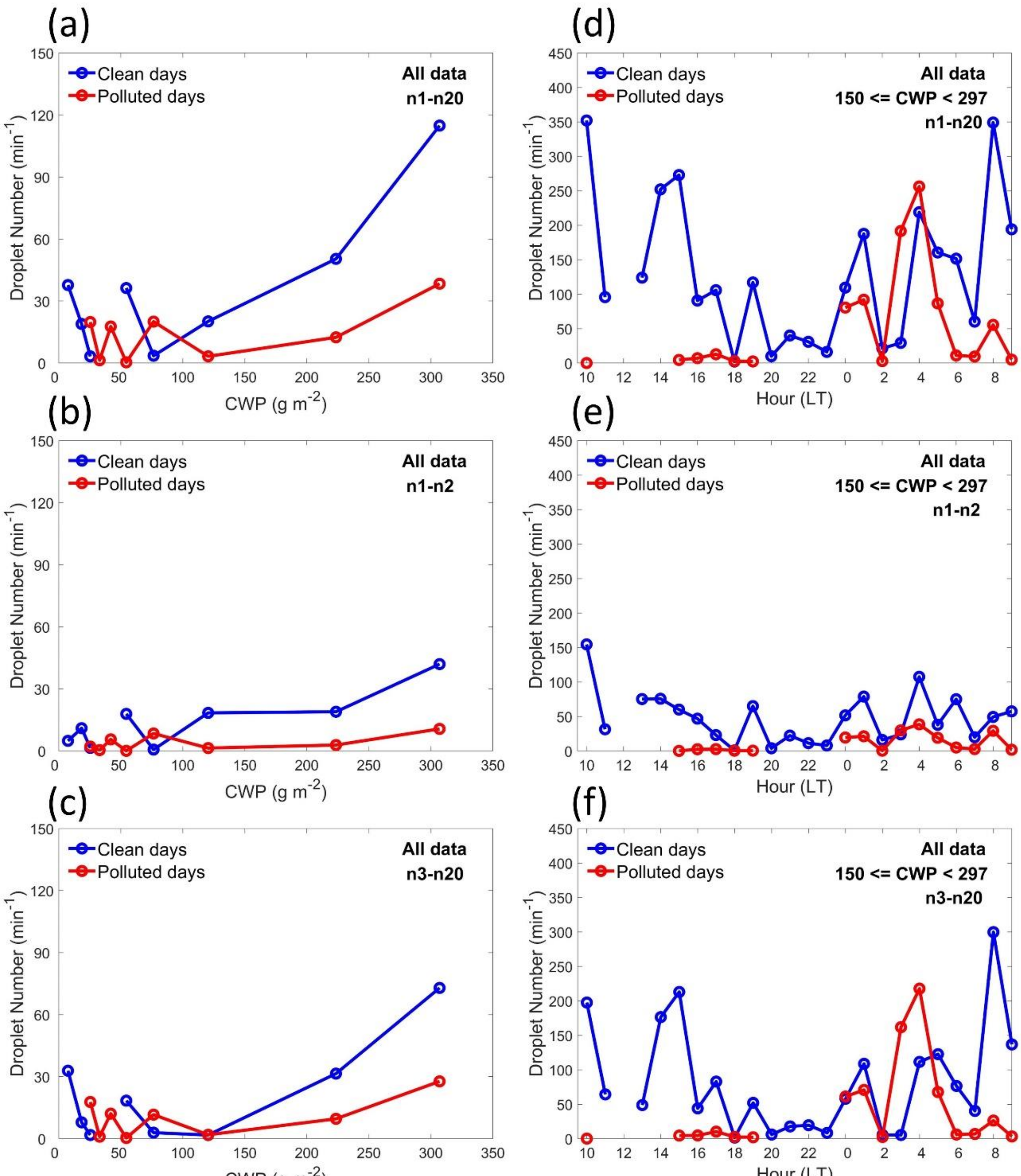



Figure R2: Multiyear (2005-2017) mean droplet number for (a) all droplets, (b) drizzle drops, and (c) raindrops in different CWP groups calculated for clean and polluted days. Hourly trend of mean droplet number for (d) all droplets, (e) drizzle drops, and (d) raindrops calculated for clean and polluted 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.

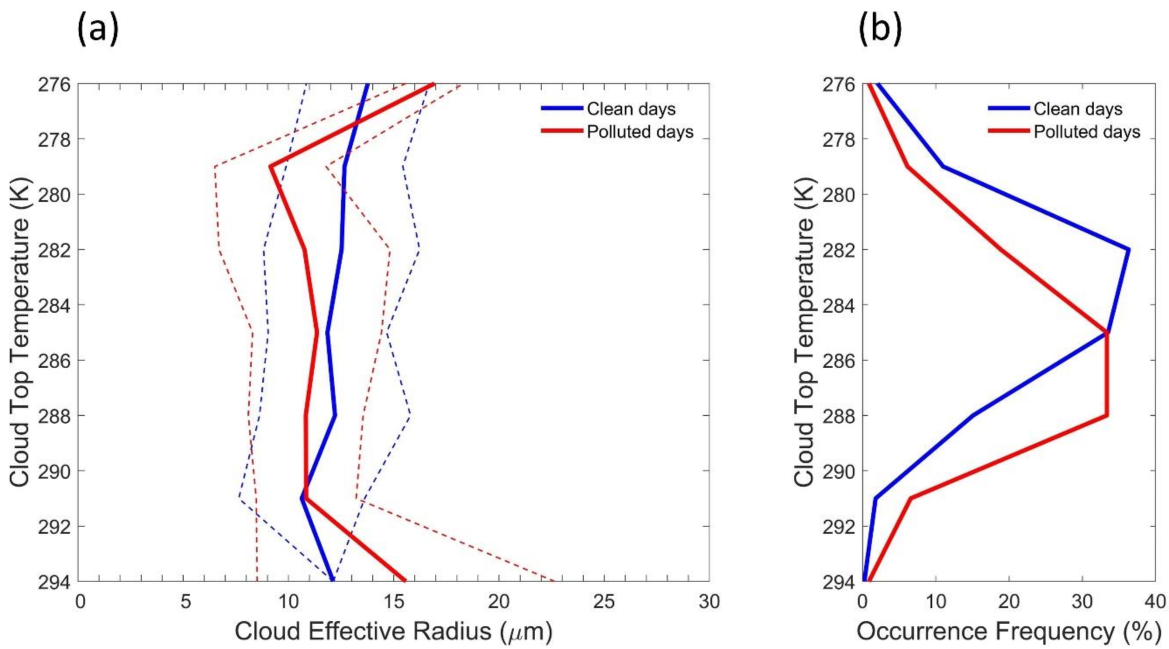



Figure 8: Multiyear (2005–2017) (a) cloud top temperature (CTT)-cloud effective radius (CER) relationship. Plotted are the mean (solid line) and one standard deviation (dashed line) of the CER for each 3 K interval, and (b) Frequency of occurrence of the CTT. Clean and polluted days are depicted with 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 ($150 \leq CWP < 297$), 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 ($\leq 1$ 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 ≤ 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.

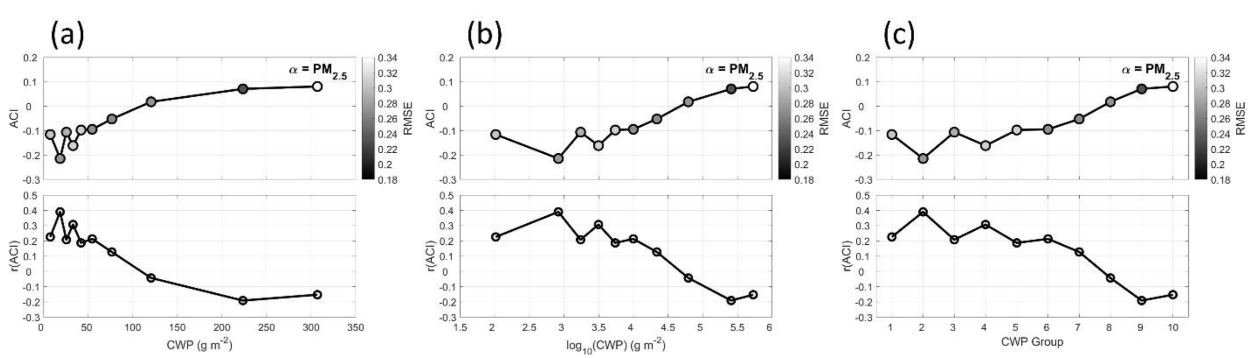
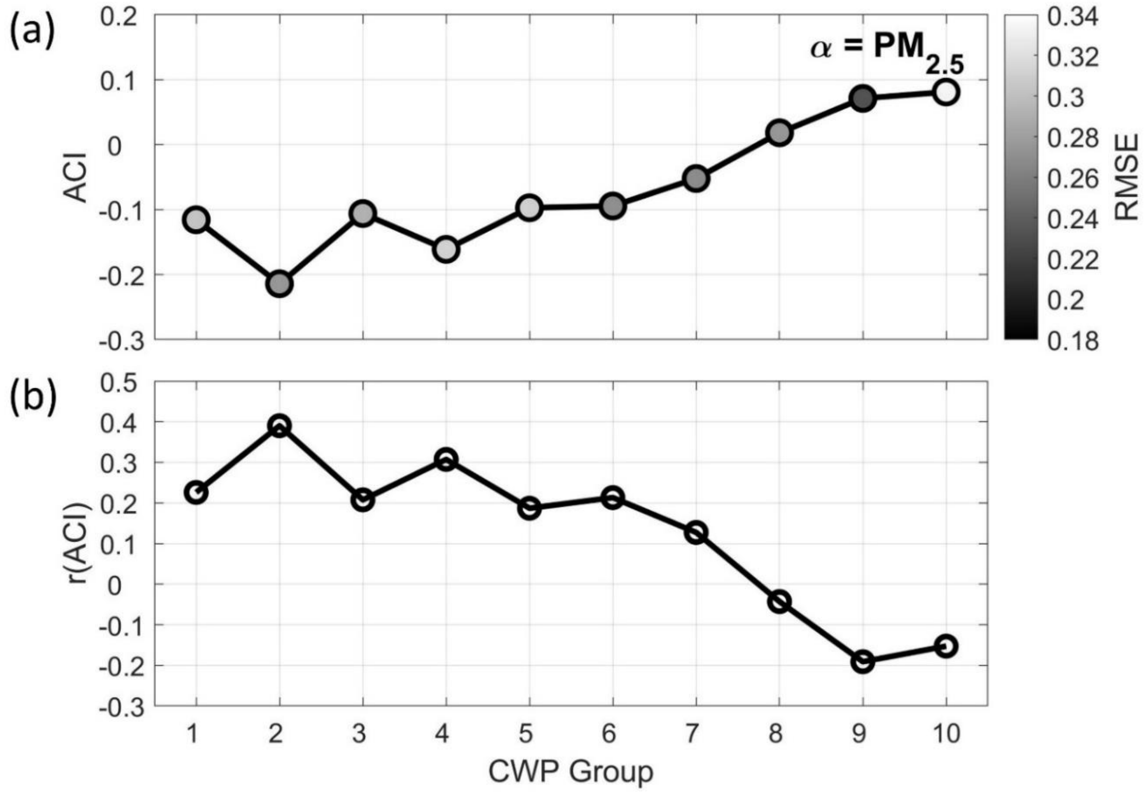



Figure R3: Aerosol cloud interaction estimated values vs. three different CWP variables (a) CWP, (b) logarithmic CWP, and (c) CWP group number.




220 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.

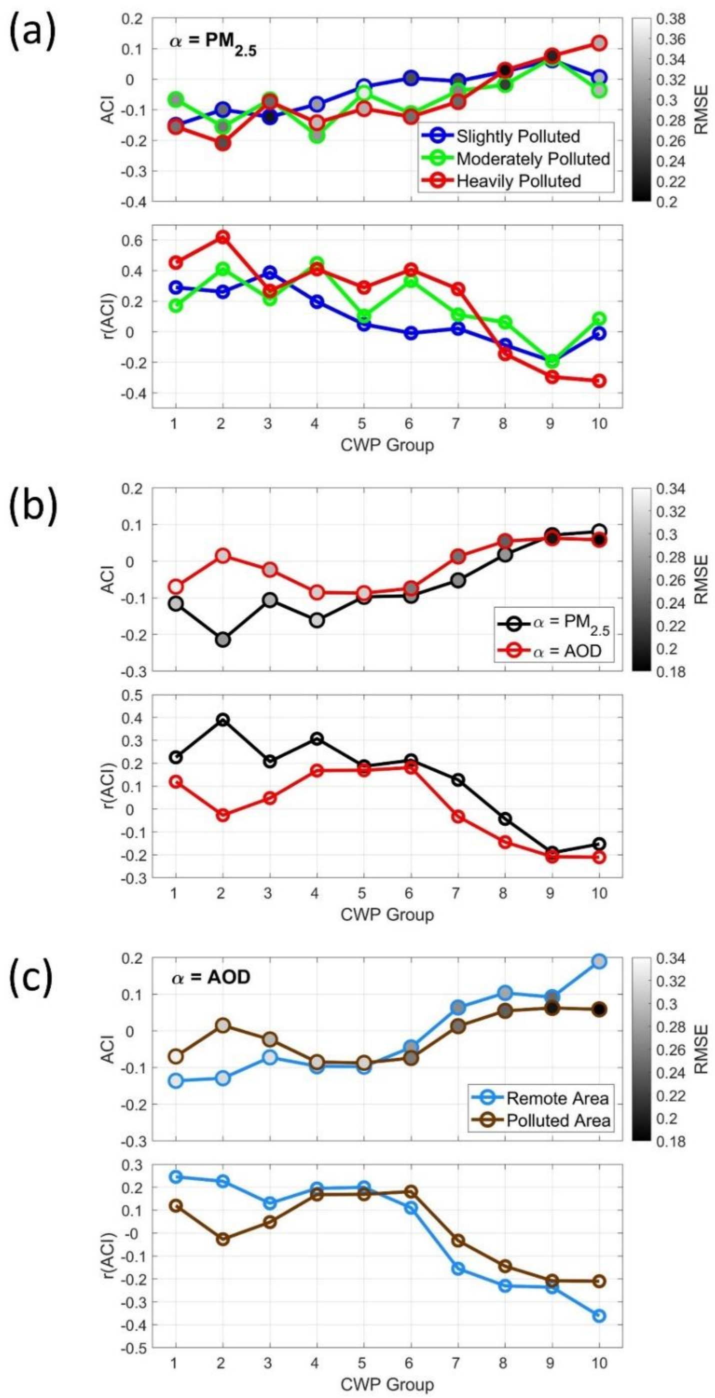



Figure 9: Multiyear (2005–2017) ACI values with the RMSE (shaded) and the correlation coefficient among (a) different polluted levels, (b) different aerosol proxies, and (c) different polluted condition areas.

**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.

# Response to the reviewer 2 comments for "Aerosol impacts on warm-cloud microphysics and drizzle in a moderately polluted environment" by Chen et al.

**Anonymous Referee #2**

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.

The authors present a nice, if perhaps a little over-extensive, study looking at in situ and some satellite measurements in an urban and complex setting. While the analysis presented here in some cases is not new, the data analysis of in situ data is hard and different and the analysis warrants publishing to add to our growing knowledge of aci.

We appreciate the reviewer for recognizing the value of this work. Specific points raised by the reviewer have been carefully considered and addressed in the following replies. In particular, much of the related revisions are focused on aerosol and cloud properties used in ACI and the discussion of the relevant results. We have addressed each comments in the sections below and made revisions to the manuscript accordingly.

I find some of the discussion of adjustments overly assertive of causality, which the authors cannot show empirically. These regions need to be trimmed to report on findings without asserting a causal connection, or the authors should perform modelling of the region where they can make some advances to understanding the direction of causality in what their observations are doing.

We thank the reviewer for pointing out this concern. We also agree that a modeling study may enhance our knowledge of the causality; however, this would extend us beyond our current capacity. Instead of a modeling component, we have revisited our observational data, with particular focus on CER-CTT statistics and raindrop size distribution analysis, allowing us to obtain a process-level insight into aerosol impacts on drizzle and precipitation. In the revised manuscript, we have added two figures about aerosol effects on precipitation. Figure 10 shows the multiyear (2005-2017) JWD sample number (days), mean droplet number per minute and the differences between polluted and clean days of the mean droplet number in each bin. The droplet number in the n2 bin was significantly lower on polluted days, indicating less drizzle in that condition. Fig. 11 shows differences between polluted and clean days in the percentage of the cumulative droplet number distribution for (a) all data and (b) data with precipitation less than or equal to 1 mm $h^{-1}$. The results using all data are similar with Fig. 10c; the droplet numbers appear lower for the smaller raindrop bins (≤ n5) on polluted days compared to clean days, and higher for the larger raindrop bins (> n5) (Fig. 11a). When precipitation is lower than or equal to 1 mm $h^{-1}$ (i.e. light rain), abundant CCN drives raindrops towards smaller drop sizes, effectively increasing the number of drizzle drops (Fig. 11b).

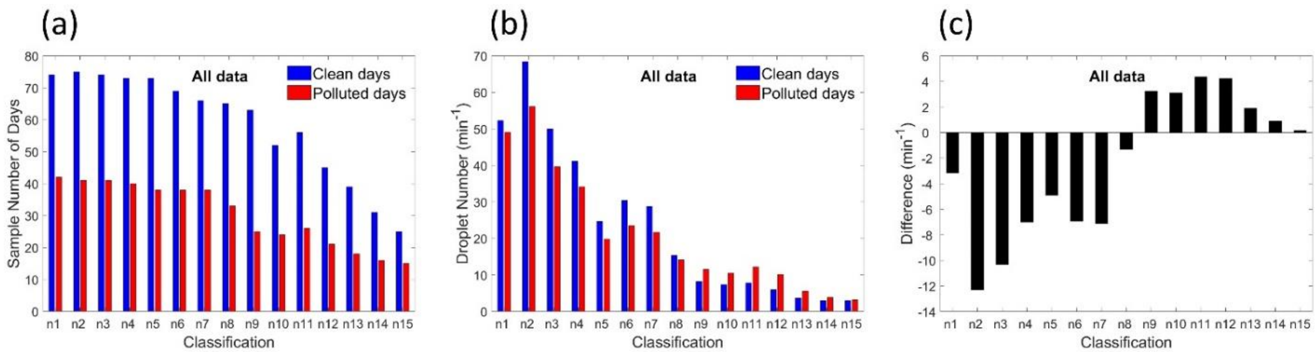



Figure 10: Multiyear (2005-2017) (a) JWD sample number of days in each raindrop size bin, (b) mean droplet number per minute for clean and polluted days and (c) The differences in the mean droplet number between polluted and clean days. nX reflects different raindrop size bins. The droplet size for n1 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.

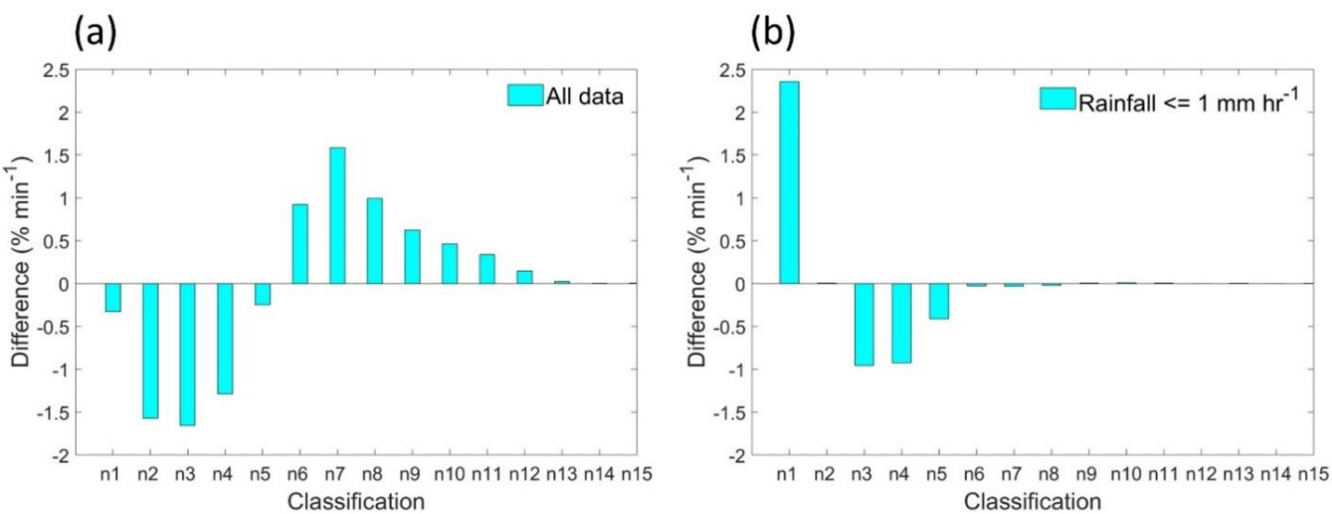



Figure 11: Multiyear (2005-2017) differences between polluted and clean days as percentages of the cumulative droplet number distribution for (a) all data and (b) the data with precipitation less than or equal to 1 mm $h^{-1}$. nX reflects different raindrop size bins as listed in Fig. 10.

In addition, we rewrote some paragraphs of findings by adding references rather than asserting a causal connection. The summary paragraph in Sect. 3.4 is rephrased as (lines: 297-303):

Although the existence of an aerosol effect on cloud lifetime is still widely disputed (Small et al., 2009; Stocker, 2014), our preliminary results show that precipitation might be suppressed and delayed under high aerosol loading. Combined with the results from Sect. 3.2, the process in the aerosol-cloud-precipitation interactions is consistent with the cloud lifetime effect. The presence of aerosols enhances the concentration of condensation nuclei under a fixed water content, which increases the cloud droplet number, redistributes cloud water to more numerous and smaller droplets, reducing collision–coalescence rates, which in turn suppresses precipitation and delays rainfall occurrence (i.e. the cloud lifetime effect (Albrecht, 1989; Pincus and Baker, 1994; Lohmann and Feichter, 2005)).

And a portion of the conclusions has been rephrased as (lines: 313-331):

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 ($150 \leq CWP < 297$), 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 ($\leq 1$ mm $h^{-1}$) category, high concentration of aerosols drove raindrops towards smaller droplet sizes and increased the appearance of drizzle drops.

While I acknowledge that many studies utilize CER to calculate aci, I would suggest using $N_d$, which the authors have already calculated to provide a complimentary calculation that may be more relevant to more recent studies.

Thank you for the suggestion. We agree with the reviewer's insight that cloud droplet number concentration ($N_d$) calculation may be more relevant. Grosvenor et al. (2018) indicated that $N_d$ is of central interest to improve the understanding of cloud microphysics and for quantifying the effective radiative forcing by aerosol-cloud interactions. However, current standard satellite retrievals do not operationally provide $N_d$. It can be inferred from retrievals of cloud optical depth (COD), cloud droplet effective radius (CER) and cloud top temperature, but errors propagated from passive retrievals of COD and CER will generate uncertainties in the subsequently derived $N_d$ (Grosvenor et al., 2018); thus, we currently retain the calculation of ACI by using CER.

The authors may also wish to say a few words about why $PM_{2.5}$ may be a good CCN and need to address near-cloud aerosol swelling in the text, which makes the direction of causality even more difficult to infer. The use of the rain size distribution is a good way to approach this problem.

Thank you for the suggestion. In the revised manuscript, we have added a relevant sentence to specify the $PM_{2.5}$ characteristics that we considered for using it as a suitable proxy of CCN (lines: 110-113):

The composition of $PM_{2.5}$ in East Asia is usually dominated by carbonaceous species and water soluble ions, including $SO_4^{2-}$, $NH_4^+$, and $NO_3^-$ (Xu et al., 2012), which are important in determining the hygroscopicity of aerosols (Shen et al., 2009). Thus, based on these suitable characteristics and the lack of measured CCN in this study, we used $PM_{2.5}$ as a proxy for CCN concentrations.

Li et al. (2017) used $PM_{2.5}$ measurements to represent aerosol loading under cloudy conditions and showed significant negative relationships between cloud droplet effective radius (CER) and $PM_{2.5}$. Large-scale measurements of cloud condensation nuclei (CCN) are difficult to obtain on a routine basis, whereas aerosol optical quantities are more readily available (Liu and Li, 2014). However, AOD is not available under cloudy conditions, and AOD cannot represent the aerosol concentrations at the bottom of the cloud, leading to uncertainties in aerosol-cloud-precipitation interaction studies (Liu et al., 2020). Thus, hourly in-situ measurements, such as $PM_{2.5}$, are an alternative choice to estimate aerosol loading under cloudy conditions.

Aerosol swelling in high humidity cloudy environments (Clarke et al., 2002) is a possible reason behind the large uncertainties in aerosol-cloud interaction (ACI) studies using satellite retrievals (Liu et al., 2018). To address the near-cloud aerosol swelling in the text, we now reference the analysis methodology of Rosenfeld (2000), and have replotted the mean and one standard deviation of CER at each CTT bin in Fig. 8 as below. We defined the clean/polluted days by using surface $PM_{2.5}$ data, and then displayed the CTT-CER relationship and the occurrence frequency (%) of the CTT in CWP group 9 on clean and polluted days. This avoids the effect of near-cloud aerosol swelling, because $PM_{2.5}$ observations were at the surface. Figure 8 showed that CTT between 285 and 288 K exhibited a higher occurrence frequency during polluted days, whereas clean days had a higher frequency of CTT between 282 and 285 K. These results suggest that abundant aerosols activated higher concentrations of CCN near surface, thus forming more low-level clouds with smaller cloud droplet size.

In the revised manuscript, we are able provide insights to our research questions, but there are still many uncertainties. For example, $PM_{2.5}$ is not equal to CCN, satellites cannot observe particle size distribution, and it is difficult ensure our representative aerosols concentrations are present in the cloud. We appreciate that the reviewer suggested such a helpful addition; analysis of rain droplet size distribution provided us another independent verification, which made us more confident in our results.

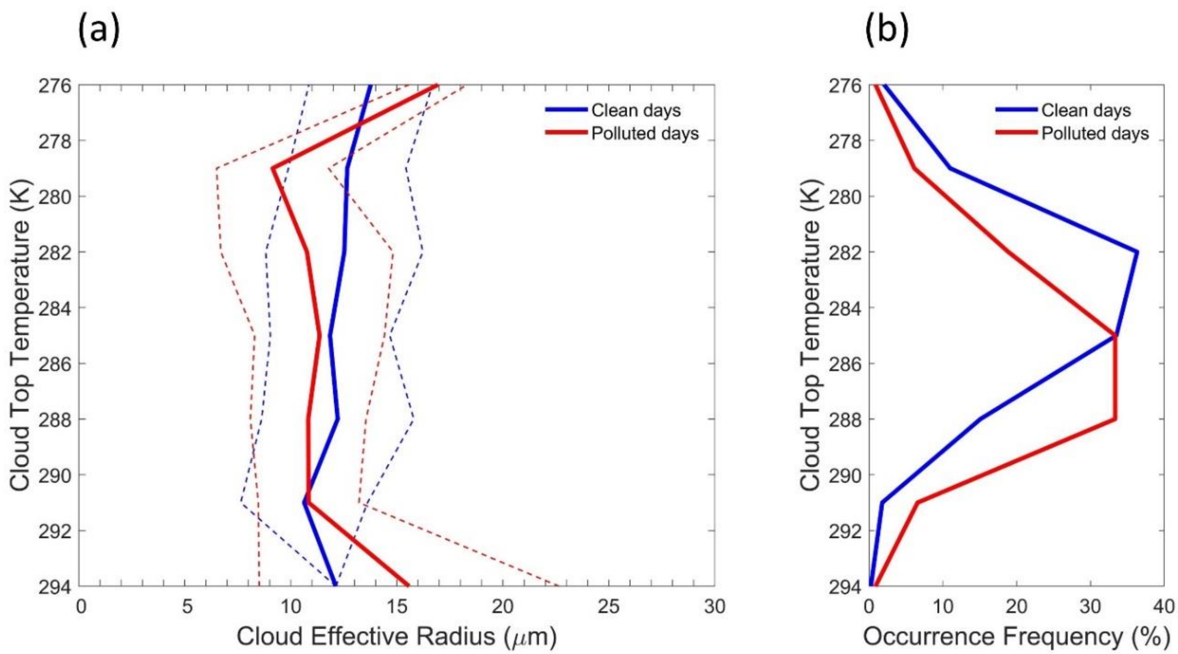



Figure 8: Multiyear (2005–2017) (a) cloud top temperature (CTT)-cloud effective radius (CER) relationship. Plotted are the mean (solid line) and one standard deviation (dashed line) of the CER for each 3 K interval, and (b) Frequency of occurrence of the CTT. Clean and polluted days are depicted with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

Another way the authors might want to consider looking at this is performing the same analysis in their paper, but instead of sorting clean/polluted sorting by atmospheric advection from the east or west. This might reveal the underlying meteorological signal that will covary with aerosol. This result can be used to say ‘on days when the dominant weather pattern is such, but there is unusually little aerosol then the clouds do this’.

We plotted the wind rose diagrams of wind speed, relative humidity, and $PM_{2.5}$ concentration at Pingzhen station from Oct.15 to Nov. 30, 2005-2017 (Fig. R1). The prevailing wind was northeast, the highest occurrence frequency for wind speed was about 4 m $s^{-1}$, relative humidity was between 70% and 90%, and the $PM_{2.5}$ concentration was below 50 μg $m^{-3}$.

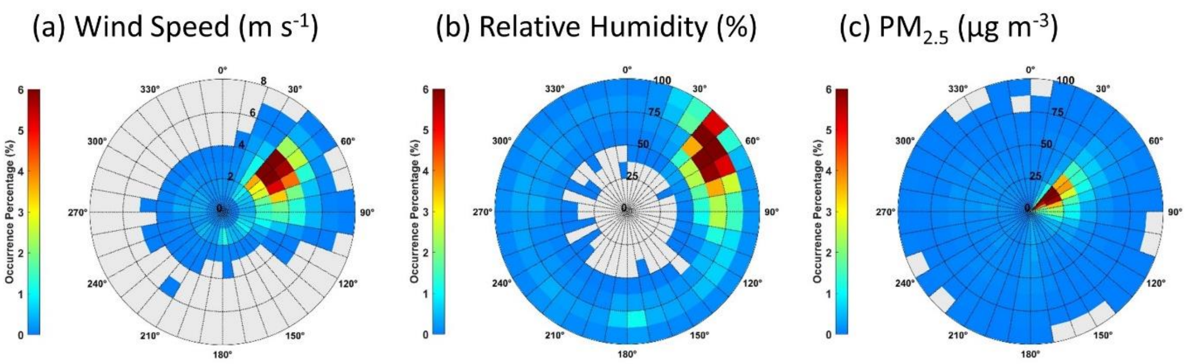



Figure R1: Multi-year (2005-2017) wind rose diagrams of (a) wind speed, (b) relative humidity, and (c) $PM_{2.5}$ concentration at Pingzhen station during Oct. 15 to Nov. 30.

To discuss the effect of atmospheric advection, we replotted Fig. 7 from the manuscript, but sorted by east wind and west wind (Fig. R2). If the most frequently occurring daily wind direction was between 0°-180°, it was defined as east wind; vice versa, it was defined as west wind when it was between 180°-360°. The difference in cloud microphysical parameters between east wind and west wind over the main research area (24.6°–25.2° N, 120.9°–121.5° E) was then calculated (Fig. R2). The East-West difference in COT, CER, CF, and CTT was -3.02, +0.47 μm, +0.01, and −1.50 K, respectively. Overall, the samples on west wind days were less available, although 13-years data were included. Thus, for the current manuscript, we decided to retain the analysis of clean vs. polluted days, but will consider a comprehensive analysis separating east vs. west air mass origins for a future work.

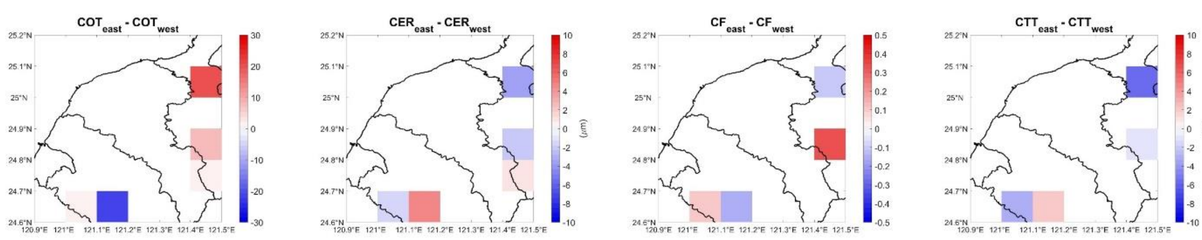



Figure R2: Multiyear (2005-2017) difference in (a) COT, (b) CER, (c) CF, and (d) CTT between east wind and west wind when considering only CWP group 9 ($150 \leq CWP < 297$). White parts are missing values.

Regarding meteorological parameters and $PM_{2.5}$ concentrations at Pingzhen station (Fig. 4), lower relative humidity, less rainfall, higher frequency of northeast wind, and lower wind speed were associated with polluted days compared with clean days. A weaker and more disorderly direction of the wind was observed on polluted days, which suggests that pollution may be associated with more stagnant conditions. However, on polluted days, the prevailing wind direction was still northeast, indicating although differences in the meteorological condition were evident between clean and polluted days, the predominant wind direction was the same. Due to the difference in meteorological conditions were between clean and polluted days, we tried to avoid any consequent impact by constraining the key cloud microphysical parameter CWP (i.e. performed analysis only on CWP group 9). We indeed understand the reviewer's concern, so we plotted Fig. R2 and attempted to clarify the effect of meteorology. However, due to a prevailing northeast wind throughout the sampling period, the samples associated with west wind days were not sufficient to represent a robust result.

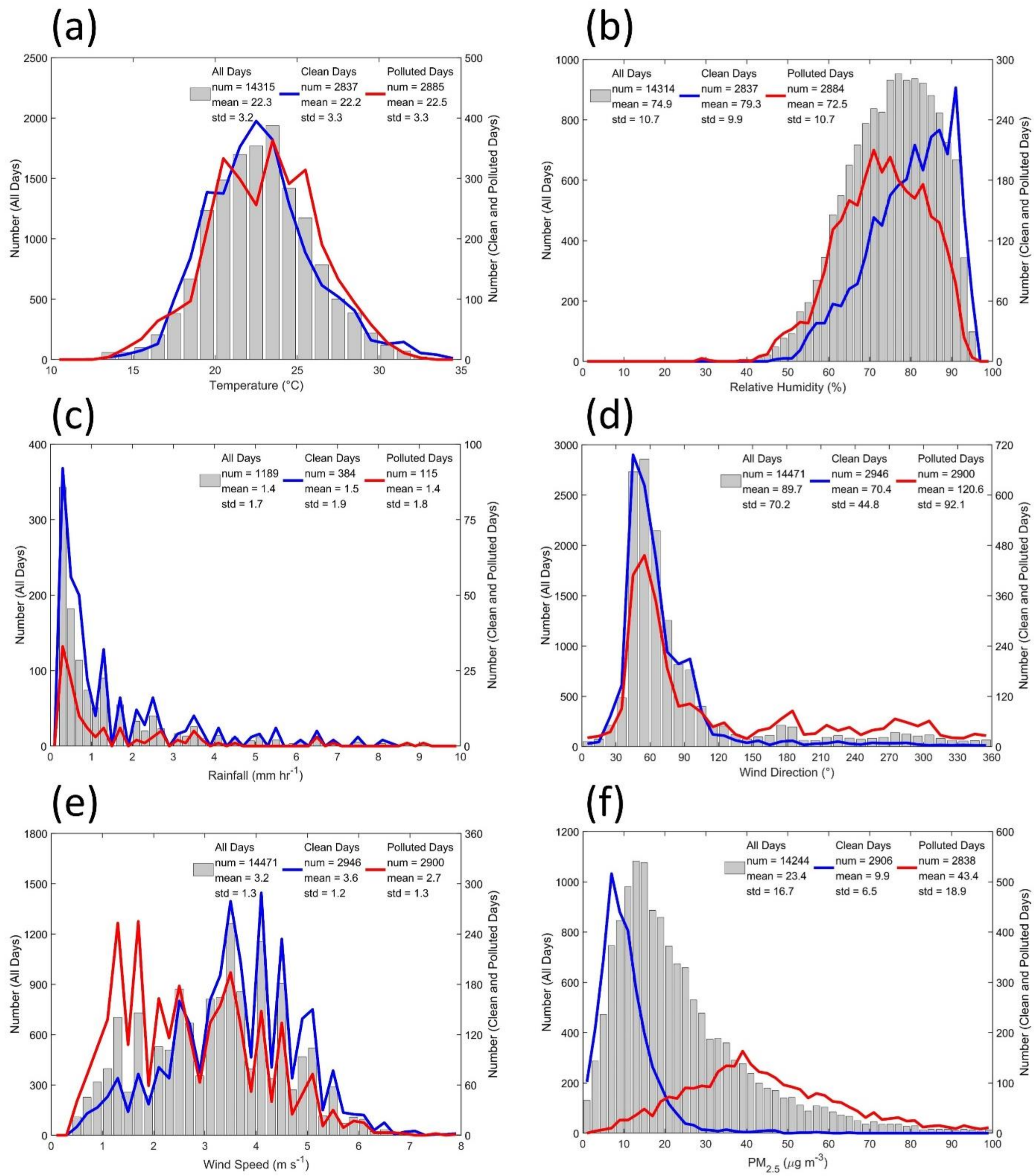



Figure 4: The distribution of (a) temperature, (b) relative humidity, (c) rainfall, (d) wind direction, (e) wind speed, and (f) $PM_{2.5}$ hourly data from Pingzhen station from Oct. 15 to Nov. 30, 2005–2017. The gray bars are the distribution of all valid observations, the blue lines represent the clean days and the red lines represent the polluted days.

P1 L15: I am not sure what this sentence is getting at- is the human activity causing low cloud?

Thank you for pointing out the confusion. We want to describe, based on a long-term analysis, that there are more low-level clouds and high AOD in northwestern Taiwan than northeastern Taiwan. In the revised manuscript, the sentence has been corrected as (lines 15-17):

Our results indicated that northwestern Taiwan, which has several densely populated cities, is dominated by low-level clouds (e.g. warm, thin, and broken clouds) during the fall season.

P2 L17: You should discuss spurious correlation between AOD and cloud properties as shown in (Christensen et al. 2017; Twohy et al. 2009).

Thank you for the suggestion. We have added discussion of spurious correlation in the text at (lines: 46-49):

Likewise, Twohy et al. (2009) and Christensen et al. (2017) reported spurious correlations between AOD and cloud properties using in-situ aircraft and satellite data. Despite advances in satellite-based retrievals in recent decades, obtaining robust statistical relationships between aerosols and clouds is difficult using only satellite-based observations (Christensen et al., 2017).

P2 L31: I might say weakly constrained (Bellouin et al. 2020).

The sentence has been rephrased as (lines: 60-62):

Although numerous studies have used observations and model simulations to discuss the indirect effects of aerosols, the interaction mechanism between aerosols and clouds remains weakly constrained (Bellouin et al., 2020) in the global climate system.

P3 L3: What does largely dominant mean? Relative to what?

Thank you for pointing out the confusion. In the revised manuscript, the sentence has been revised as (lines: 66-68):

Furthermore, Giorgi et al. (2003), using a coupled regional chemistry–climate model, found that **aerosol indirect effects were largely dominant over direct effects** in inhibiting precipitation in East Asian climates.

P3 L6: It seems like it might be good to discuss this in the context of the current synthesis report on aci (Bellouin et al. 2020).

Thank you for the suggestion. We have added discussion referencing the suggested reference as (lines: 71-74):

These studies have demonstrated a significant correlation between aerosols and cloud microphysics and the indirect effect of aerosols on regional precipitation. However, the aerosol type, concentration, and characteristics vary by region. **Moreover, the uncertainty on radiative forcing, especially via the impact from clouds remains large in Earth's radiation budget (Bellouin et al., 2020).**

P4 L13: So AOD was only retrieved when AOD was visible? It seems like all periods with cloud should be zeroed out since there might be AOD below cloud that is not being counted.

AOD is not available under cloudy conditions, and AOD cannot represent the aerosol concentrations at the bottom of the cloud. To compensate for this limitation, densely available surface $PM_{2.5}$ data in the study domain was used to resolve this condition, although this assumes that the measured $PM_{2.5}$ concentrations are representative of that within the cloud. While we have $PM_{2.5}$ observations from land-based stations, we relied solely on satellite data for aerosol proxy (i.e. AOD) levels within grid cells over the sea. In this research, to ensure the integrity of the data, satellite data, with a resolution of cloud properties at 1 and 5 km and aerosol properties at 10 km, were interpolated to a coarse resolution of 0.1° × 0.1°. These limitations lead to uncertainties in the study of aerosol-cloud-precipitation interaction that we are currently unable to improve; thus, there is room for improvement in future works.

P4 L19: What is this based on? Afternoon aerosol should be able to affect afternoon clouds.

Thank you for pointing out the confusion. In the revised manuscript, the sentence has been corrected as (lines: 119-121):

**Fine particles were assumed well-mixed throughout the PBL during daytime (Maletto et al., 2003).** $PM_{2.5}$ data between 10:00 and 14:00 were averaged as a measure of daily $PM_{2.5}$ concentrations for comparison with Aqua satellite data (overpass time is approximately 13:30 local time).

P5 L1: This would be more reliably at a constant CWP if cloud droplet number concentration ($N_d$) was used instead of CER and binning by CWP (Grosvenor et al. 2018). Any inferred aci will be a function of binning decisions.

We agree with the reviewer's insight that a constant CWP would be more reliable if we used cloud droplet number concentration ($N_d$) instead of CER. Grosvenor et al. (2018) indicated that $N_d$ is of central interest to improve the understanding of cloud physics and for quantifying the effective radiative forcing by aerosol-cloud interactions. Current standard satellite retrievals do not operationally provide $N_d$, but it can be inferred from retrievals of cloud optical depth (COD) cloud droplet effective radius (CER) and cloud top temperature.

However, errors propagated from passive retrievals of COD and CER will generate uncertainties in the subsequently derived $N_d$. The CER uncertainties are likely to have a larger impact than COD errors due to the larger sensitivity of $N_d$ to CER. Retrievals based on MODIS and other instruments employ bispectral algorithms for retrieving COD and CER (Nakajima & King, 1990), whereby these quantities are estimated using reflectances from both a nonabsorbing visible wavelength (denoted here as $R_{vis}$) and an absorbing shortwave infrared wavelength ($R_{SWIR}$). To observe $R_{vis}$, the instrument uses the 0.65-μm channel over land and the 0.86-μm channel over the ocean. Since surface albedo errors can be large, it is worth discussing them further, although we note that the uncertainties examined above in Platnick et al. (2017) were over the land, where MODIS surface albedo uncertainties are likely to be much higher than over the oceans (Bréon & Doutriaux-Boucher, 2005; King et al., 2004; Rosenfeld et al., 2004) since the surface albedo over land is much more variable than over the ocean (Grosvenor et al., 2018).

In summary, we agree with the reviewer's insight that it may be more reliable if cloud droplet number concentration ($N_d$) was used instead of CER, but to avoid the large uncertainties associated with derived $N_d$, we currently maintain the calculation of ACI by using CER.

P7 L7: This is a nice comparison to previous studies. Please consider summarizing in a figure.

Thank you for the suggestion. We have collected this information in Table 2 as below:

Table 2: ACI values from the literature in comparison to this study.

| **Study** | **ACI values** | **Sources** | **Region** |
|---|---|---|---|
| Feingold et al., 2003 | 0.02-0.16 | ground-based remote sensors | Oklahoma, United States |
| Kim et al., 2008 | 0.04-0.17 | ground-based remote sensors | Oklahoma, United States |
| McComiskey et al., 2009 | 0.04-0.15 | ground-based remote sensors | California, United States |
| This study | 0.07 in CWP group 9 ($150 \leq CWP < 297$) | satellite and surface observations | Northern Taiwan |

P7 L13: This is not a robust piece of analysis. Differing $PM_{2.5}$ is likely a function of atmospheric state (air masses moving from the west for instance) and this is likely to do more to CF and COT than aci.

Thank you for pointing this out. We agree with the reviewer's insight of impact from atmospheric state. As discussed above for Fig. R2, the difference in cloud microphysical parameters between east wind and west winds affecting the main research area were calculated. The samples on west wind days were too few even though 13 years of data were included. Although meteorological conditions were different between clean and polluted days, we attempted to avoid an effect from meteorology by constraining the key cloud microphysical parameter CWP (i.e. performed analysis on CWP group 9 only). We indeed understand the reviewer's concern, and tried to clarify the effect from meteorology. Owing to the prevailing northeast wind, the samples on west wind days were not sufficient to represent a robust result. Currently, in the revised manuscript, we added a reference to discuss the meteorological impact, and the paragraph has been revised as (lines: 201-203):

While the positive CF value difference may have been due to higher aerosol loading, the atmospheric condition may have contributed as well. For instance, Saponaro et al. (2017) showed that CF is more sensitive to lower troposphere stability (LTS) than other cloud variables (i.e. CER, CTT, and COT).

P8 L6: Please comment on the unintuitive diagnosed stronger aci in more polluted clouds. A lot of studies point to stronger aci in more pristine clouds (Carslaw et al. 2013). Again, this may be a function of binning, which is also going to select for clouds in an atmospheric regime.

Thank you for the comment. The ACI calculation is dependent on how the environmental factors are constrained (i.e., fixed CWP) and on the data binning. In our results, the differences across three polluted levels are relatively small; ACI values were 0.08, 0.07, and 0.06 for heavily, moderately, and slightly, respectively, when considering only CWP group 9 ($150 \leq CWP < 297$) data. This implies that ACI changes between different polluted levels may not have been significant as long as the clouds contained a certain amount of CCN. Without in-situ measurement, we are not able to definitively verify our results. We are planning a more comprehensive measurement study to quantify the ACI index in our future works.

P9 L4: Or the cleaner days could be occurring because of rain scavenging aerosol. Unfortunately, in an empirical study such as this you can't make causal statements. However, the high temporal resolution of ground data used here might allow for some sort of time evolution analysis that could show causality.

Thanks for the comments. We agree with the reviewer's insight and have avoided an overly assertive statement of causality by rephrasing the sentence as (lines: 257-259):

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.**

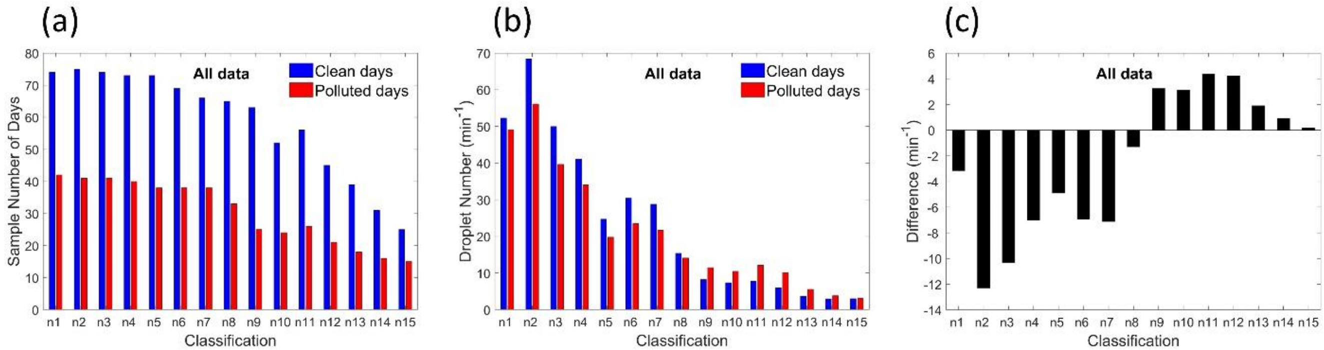



Figure 10: Multiyear (2005-2017) (a) JWD sample number of days in each raindrop size bin, (b) mean droplet number per minute for clean and polluted days and (c) The differences in the mean droplet number between polluted and clean days. nX reflects different raindrop size bins. The droplet size for n1 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.

As the reviewer pointed out, it is overly assertive to make causal statements in an empirical study. To further improve this part of the analysis, we traced back the timeframe to 00:00 as shown in Fig. R3. $PM_{2.5}$ data were averaged from 10:00 to 14:00 as daily $PM_{2.5}$ (orange box). Therefore, the rainfall data from 00:00 to 10:00 represents the pre-setting period of clean/polluted days. For the clean days, rain scavenging occurred in the morning, especially during the period of 05:00-10:00. The rainfall and $PM_{2.5}$ concentrations were both at low values from 10:00-14:00. The rainfall variability during clean days can be considered as the typical pattern without aerosol effects. For polluted days, there was scarcely rainfall occurring before 10:00. After 10:00, when daily $PM_{2.5}$ was higher, high aerosol concentrations led to higher concentrations of CCN, produced more liquid particles and inhibited the cloud droplet size (as the Sect. 3.2 shown). Precipitation started early in the night (i.e. 12-16 hrs after the $PM_{2.5}$ averaging period). Synthesizing the results mentioned above, under a fixed CWP ($150 \leq CWP < 297$), a higher aerosol concentration redistributes cloud water to more numerous and smaller droplets, reducing collision–coalescence rates, which in turn suppress precipitation and constrain the time of rainfall occurrence.

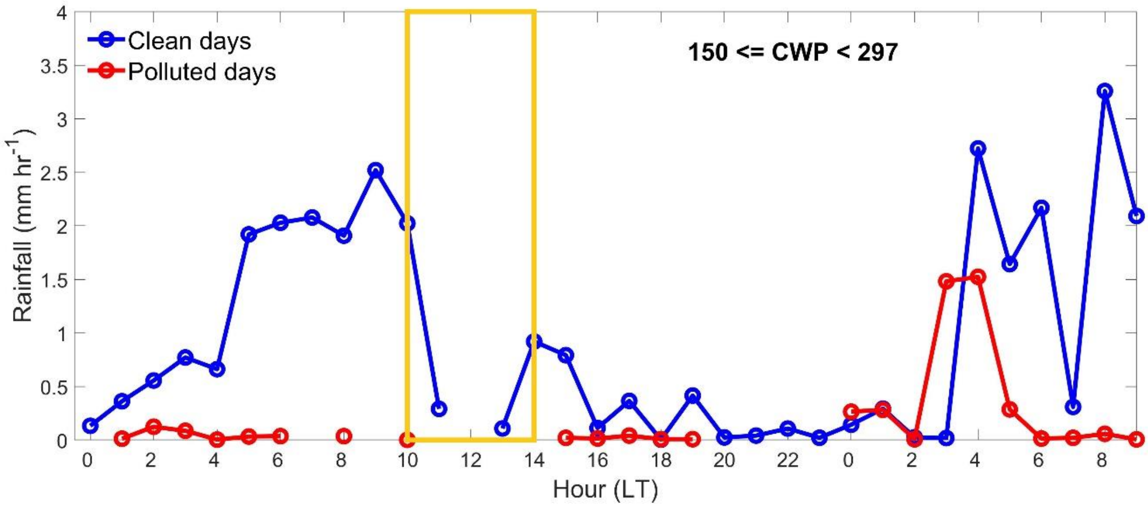



Figure R3: Time series of average hourly-rainfall rate calculated for clean and polluted days when considering CWP group 9 ($150 \leq CWP < 297$) only. $PM_{2.5}$ data were averaged from 10:00 to 14:00 as daily $PM_{2.5}$ (orange box) and rainfall analyses were performed from 00:00 of that day (i.e. before the $PM_{2.5}$ averaging period) to 10:00 of the following day.

P9 L22: Please note that precipitation reduction is often a function of model parameterization.

The sentence has been rephrased as (lines: 284-286):

A modeling study (Huang et al., 2007) revealed that the second indirect effect of aerosols (a large number of small droplets are generated by enhanced aerosols and reduce the precipitation efficiency) significantly reduces fall and winter precipitation from 3 % to 20 % across East Asia, although it was **dependent on the auto-conversion scheme assumed.**

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# Aerosol impacts on warm-cloud microphysics and drizzle in a moderately polluted environment

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**Abstract.** Climate is critically affected by aerosols, which can alter cloud lifecycles and precipitation distribution through radiative and microphysical effects. In this study, aerosol and cloud property~~ies~~ datasets from MODIS onboard the Aqua satellite, and surface observations, including aerosol concentrations, raindrop size distribution, and meteorological parameters, were used to statistically quantify the effects of aerosols on low-level warm cloud microphysics and drizzle over northern Taiwan during fall seasons (from October 15 to November 30 of 2005–2017). Our ~~R~~results indicated that ~~clouds in~~ northwestern Taiwan, which ~~with active human activity~~ has several densely populated cities, is dominated by low-level clouds (e.g. warm, thin, and broken clouds) during the fall season. The observed effects of aerosols on warm clouds indicated aerosol indirect effects, i.e.~~;~~ increased~~ing~~ aerosol loading caused a decrease in cloud effective radius (CER), an increase in cloud optical thickness, an increase in cloud fraction, and a decrease in cloud top temperature under a fixed cloud water path. ~~A q~~Quantitatively, ~~value of~~ aerosol–cloud interactions ($ACI = \partial \ln CER/\partial \ln \alpha$, changes in CER ~~depend on~~ relative to changes in aerosol~~s~~ amounts) were ~~calculated to be~~ 0.07 for our research domain~~. ACI values~~, and varied between 0.09 and 0.06 in the surrounding~~ clean~~ remote (i.e. ocean) and ~~heavily~~ polluted (i.e. land) areas, respectively, ~~which~~ indicating~~ed that~~ aerosol indirect effects were stronger ~~more sensitive~~ in the remote~~clean~~ area. ~~Analysis of~~ From the raindrop size distribution analysis, ~~observations during~~ high aerosol loading resulted in a decreased frequency of drizzle events, redistribution~~ed~~ of cloud water to more numerous and smaller droplets, and reduced collision-coalescence rates. However, ~~in the scenario of light precipitation~~during light rain ($\leq$1 mm h$^{-1}$), high aerosol concentrations dro~~i~~ve raindrops towards smaller droplet sizes and increased the appearance of drizzle drops. This study used long-term surface and satellite data to determine aerosol variations in northern Taiwan, effects on the clouds and precipitations, and ~~applications to~~ observational strategies~~y planning~~ for future research on aerosol–cloud–precipitation interactions.

## 1 Introduction

Since the industrial revolution, the quantity of aerosols produced by human activities has increased significantly~~. Aerosols are most concentrated in~~, with the strongest aerosol emissions from areas with frequent industrial activities or~~and~~ high biomass burning ~~because of the short lifetime of aerosols~~ (Textor et al., 2006). The effect of aerosols on climate is recognized as significant (Charlson et al., 1992; Kiehl and Briegleb, 1993; Penner et al., 2001; Ramanathan et al., 2001; Ramaswamy et al., 2001) albeit complex. Aerosols can alter cloud properties ~~and the~~ with subsequent ~~adjustments~~ impacts on climate, ~~also known as the~~ i.e. aerosol indirect effect (Warner and Twomey, 1967; Twomey, 1974; Albrecht, 1989; Lohmann and Feichter, 2005). The responses of convective and boundary layer clouds contribute to the spread of global cloud feedbacks in general circulation models (GCMs), with ~~a~~ dominate~~nt role of~~ the inter-model differences ~~in the response of low-level clouds~~ (Bony et al., 2006). ~~The concentration of aerosol particles and cloud condensation nuclei (CCN) provides a valuable link between aerosol and cloud. Aerosols can alter warm cloud characteristics through radiative and microphysical effects, which has a substantial effect on climate. However~~ Furthermore, studies have demonstrated that ~~the global model~~ GCMs significantly overestimate~~s~~ the frequency of drizzle (Stephens et al., 2010), which brings into question the accuracy of aerosol–cloud interactions (ACI)s in models. Therefore, observational studies of aerosol and cloud microphysical properties are crucial for clarifying the relationship between aerosols and the microphysical process of clouds and evaluating the accuracy of model simulations.

Jones et al. (2009) emphasized that ACI should be explored at the regional scale, because the aerosol type, concentration, and meteorological conditions differ depending on the area. Numerous studies have used the aerosol concentration and cloud droplet size to investigate ACIs at global or regional scales. A negative correlation between aerosols and cloud drop size has been observed in global (Bréon et al., 2002; Myhre et al., 2007; Nakajima et al., 2001) and regional scale (Costantino and Bréon, 2010; Ou et al., 2012) studies. Sekiguchi et al. (2003) and Grandey and Stier (2010) have used global satellite data and identified different correlations (positive, negative, or weak) between aerosol optical depth (AOD) and cloud effective radius (CER) depending on the location of the observation. Likewise, Twohy et al. (2009) and Christensen et al. (2017) reported spurious correlations between AOD and cloud properties using in-situ aircraft and satellite data. Despite advances in satellite-based retrievals in recent decades, obtaining robust statistical relationships between aerosols and clouds is difficult using only satellite-based observations (Christensen et al., 2017).

~~The effect of~~ Nevertheless, some effects from aerosols on cloud microphysics can be observed using satellite data (Krüger and Graßl, 2002; Menon et al., 2008; Rosenfeld et al., 2014; Saponaro et al., 2017; Sporre et al., 2014). With~~Using~~ satellite-based precipitation observations from the Tropical Rainfall Measuring Mission (TRMM), Rosenfeld (1999) demonstrated that aerosols derived from biomass burning suppress warm rain processes. ~~Ground observations and models have revealed a strong correlation between CER and AOD (Feingold et al., 2001; Grandey and Stier, 2010; Yuan et al., 2008).~~ Aircraft observations over the Amazon basin demonstrated decreased in-cloud droplet sizes and a delay in precipitation onset when a large quantity of aerosols entered the cloud (Andreae et al., 2004). The effects of aerosols in suppressing drizzle have been identified in field experiments on stratocumulus clouds over the northeastern Atlantic Ocean (Albrecht et al., 1995; Wood, 2005), northeastern Pacific Ocean (Lu et al., 2007; Lu et al., 2009; Stevens et al., 2003; VanZanten et al., 2005), and southeastern Pacific Ocean (Bretherton et al., 2010; Comstock et al., 2004; Wood et al., 2011). Moreover, model simulations revealed that polluted environments could suppress drizzle in warm clouds (Ackerman et al., 2004; Guo et al., 2011; Wang et al., 2011a; Wang et al., 2011b). Although numerous studies have used observations and model simulations to discuss the indirect effects of aerosols, the interaction mechanism between aerosols and clouds remains weakly constrained (Bellouin et al., 2020)~~unconstrained~~ in the global climate system.

Huang et al. (2007) used a regional coupled climate–chemical–aerosol model for the East Asia~~ region~~ and determined that the aerosol indirect effect significantly reduced precipitation in autumn and winter. Menon et al. (2002) used a global climate model to study the effects of aerosols in China and India and reported that anthropogenic aerosols increase precipitation in southeastern China but inhibited~~s~~ precipitation in northeastern China. Furthermore, Giorgi et al. (2003) used a coupled regional chemistry–climate model, found that aerosol indirect effects were largely dominant over direct effects in inhibiting precipitation in East Asian climates.~~ to assess the direct and indirect effects of anthropogenic sulphates on East Asian climates. Results indicated that aerosol indirect effects were largely dominant in inhibiting precipitation.~~ Takemura et al. (2005) used a global aerosol transport-radiation model coupled to a general circulation model and determined that the indirect effect had a strong signal in regions with large quantities of anthropogenic aerosols and cloud water.

These studies have demonstrated a significant correlation between aerosols and cloud microphysics~~. They further demonstrated an~~ and the indirect effect of aerosols on regional precipitation. However, the aerosol type, concentration, and characteristics vary by region. Moreover, the uncertainty on radiative forcing, especially via the impact from clouds remains large in Earth's radiation budget (Bellouin et al., 2020). Taiwan is an island with a high population density, a complicated topography, and a climate that ranges from tropical in the south to subtropical in the north. These characteristics result in substantially complex microphysical processes between aerosols and clouds. In this study, we aimed to systematically analyze aerosols, cloud optical properties, and precipitation characteristics by integrating satellite and surface observation data over northern Taiwan to investigate the following questions: (1) How do aerosols affect cloud microphysical properties in response to different pollution conditions? and (2) How do aerosols affect the frequency of drizzle and the change in precipitation distribution? In Sect. 2, we describe data and methodology. In Sect. 3, we present results and discussion. Findings are summarized in Sect. 4.

## 2 Data and methodology

### 2.1 Study area and time period

Our study domain, northern Taiwan, covers the area 24.5°–25.8° N and 120.8°–122.2° E (Fig. 1) and has~~with~~ a population of approximately 10 million. The emissions of this area are considered a combination of urban and industrial activities. For this area, air quality worsens~~decreases~~ in fall when precipitation is less and air masses ~~become more~~ stagnant. Moreover, the results of Huang et al. (2007) indicated~~suggested~~ that aerosol indirect effects frequently occur~~happened~~ in fall. Therefore, we chose the data period from 15 October to 30 November between 2005 and 2017 (611 days in total) to explore aerosol effects on cloud microphysics and drizzle. To remove~~prevent~~ the effect of typhoons from~~on~~ the analysis, typhoon alarm days (21–23 October 2010, Typhoon Megi) issued by the Central Weather Bureau were excluded in this study.

### 2.2 Surface measurement data

Hourly meteorological (i.e. temperature, relative humidity, rainfall, wind direction, and wind speed) and $PM_{2.5}$ concentration data collected from Taiwan EPA Pingzhen site (24.95° N, 121.20° E) and one-minute raindrop size distribution Joss–Waldvogel Disdrometer (JWD) data obtained from National Central University (NCU) (24.968° N, 121.185° E) observatory were used. The NCU and EPA Pingzhen sites are located near each other at the centre of the study domain. The $PM_{2.5}$ concentration was measured using the MetOne BAM-1020 Beta Attenuation Monitor. The JWD measures the number of rain droplets every minute by using 20 bin sizes of 0.359–5.373 mm (n1–n20: 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, 3.198, 3.544, 3.916, 4.350, 4.859, and 5.373 mm). To ensure data quality, observations were discarded when the rain rate was lower than 0.1 mm $h^{-1}$ (Greenberg, 2001; Seela et al., 2017).

### 2.3 Satellite data

Cloud and aerosol data from NASA Aqua satellite, moderate-resolution imaging spectroradiometer (MODIS) collection 6 level 2 products (MYD06 for clouds and MYD04 for aerosols) were used in this study. Data were downloaded from https://modis.gsfc.nasa.gov/data/. Data on cloud properties included cloud optical thickness (COT), CER, and cloud water path (CWP), all of which had a resolution of 1 km, as well as cloud fraction (CF), cloud-top pressure (CTP), cloud-top temperature (CTT), and cloud phase infrared (CPI), all of which had a resolution of 5 km. CWP included liquid water path and ice water path (CWP = LWP + IWP). For aerosol data, AOD with a resolution of 10 km was used. Descriptions of parameters and products are presented in Table 1. To ensure spatial resolution consistency between data sets, data were interpolated to a coarse resolution of 0.1° × 0.1°.

### 2.4 Data screening and grouping

Satellite aerosol data were not retrieved when conditions were overcast, except when aerosols were above clouds. To compensate for this limitation, densely available surface $PM_{2.5}$ data in the study domain was used. The composition of $PM_{2.5}$ in East Asia is usually dominated by carbonaceous species and water soluble ions, including $SO_4^{2-}$, $NH_4^+$, and $NO_3^-$ (Xu et al., 2012), which are important in determining the hygroscopicity of aerosols (Shen et al., 2009). Thus, based on these suitable characteristics and the lack of measured CCN in this study, we used $PM_{2.5}$ as a proxy for CCN concentrations. The spatial homogeneity of the $PM_{2.5}$ concentrations was examined based on the correlation of concentrations between the Pingzhen site and~~with~~ the 30 air quality monitoring sites in the northern~~ part of~~ Taiwan. Results indicated that correlation coefficients were higher than 0.6 and 0.8 for northern Taiwan and the research area (24.6°–25.2° N and 120.9°–121.5° E), respectively, indicating that $PM_{2.5}$ data from the Pingzhen site accurately represented the aerosol concentration~~distribution~~ over our research domain (Fig. 1).

~~Clouds and their microphysics properties in the afternoon may be affected by aerosols in the morning to noontime.~~ Fine particles were assumed well-mixed throughout the PBL during daytime (Maletto et al., 2003). $PM_{2.5}$ data between 10:00 and

14:00 were averaged as a measure of daily $PM_{2.5}$ concentrations for comparison~~to accord~~ with Aqua satellite data (overpass time is approximately 13:30 local time). Furthermore, the 20th percentile of daily average $PM_{2.5}$ data ($\leq$11.2 μg $m^{-3}$) was defined as clean days (n=123 days).~~, and the corresponding $PM_{2.5}$ concentration was 11.2 μg $m^{-3}$ for 123 days.~~ The 80th percentile of daily average $PM_{2.5}$ data ($\geq$34.6 μg $m^{-3}$) was defined as polluted days (n=121 days).~~, and the corresponding $PM_{2.5}$ concentration was 34.6 μg $m^{-3}$ for 121 days.~~ Polluted days were further divided into three groups: slightly polluted (40 days), moderately polluted (40 days), and heavily polluted (41 days) with $PM_{2.5}$ concentrations of 34.6–39.9, 39.9–52.3, and 52.3–110 μg $m^{-3}$, respectively.

A previous study ~~(Wang et al., 2010)~~ reported that the vertical aerosol distribution for the study region in autumn primarily resided ~~was mainly~~ within 2 km (Wang et al., 2010). For ACI at a local scale, clouds that occurred below 2 km ~~at the same level~~ were targeted. Therefore, only clouds with CTP ≥ 800 hPa and CPI = 1 (water cloud) were included, thereby ensuring that only warm clouds were analyzed.

To quantify ACI, the commonly used formula proposed by Feingold et al. (2001) was employed, as illustrated in Eq. (1). This equation calculates how a change in aerosols affects CER at a constant CWP.

$$\mathrm{ACI} = -\left.\frac{\partial \ln \mathrm{CER}}{\partial \ln \alpha}\right|_{CWP}, \tag{1}$$

where α represents the proxy for the quantity of aerosols, using either $PM_{2.5}$ or AOD values. Positive ACI values indicate that ~~the effect of~~ a change in CER depends on increased aerosols, and vice versa. An~~The~~ ACI value approaching 0 indicates that the relationship between CER and aerosols (i.e. aerosol indirect effect~~s~~) is not significant. The ACI calculation should be performed under a~~the~~ fixed range of CWP in Eq. (1). Therefore, the CWP population density distribution was divided into ten groups (Fig. 2), with each group representing 10 % of CWP data.

Data on the ~~precipitation~~ raindrop size distribution obtained from JWD were further processed. The daily rainfall amount was defined as the sum of precipitation from 10:00 ~~am~~ to the next day at 10:00 ~~am to investigate the consequential process of aerosol-cloud-precipitation in a day scenario by using currently available data sets~~. The AMS Glossary (Huschke, 1959) defines drizzle as very small, numerous, and uniformly dispersed water drops that may appear to float in currents. In contrast to fog droplets, drizzle falls to the ground. In weather observations, drizzle is classified as (a) “very light,” comprised of scattered drops that do not entirely wet an exposed surface regardless of the duration; (b) “light,” the rate of fall being traced to 0.25 mm $h^{-1}$; (c) “moderate,” the rate of fall being 0.25–0.50 mm $h^{-1}$; and (d) “heavy,” the rate of fall exceeding 0.5 mm $h^{-1}$. When the precipitation equals or exceeds 1 mm $h^{-1}$, all or part of the precipitation is considered rain. The threshold for rain intensity was set at 1 mm $h^{-1}$ to focus on the effect of aerosols on drizzle. Drizzle drops are conventionally 0.5 mm or less in diameter; therefore, JWD data in n1 (0.359 mm) and n2 (0.455 mm) channels were summarized as drizzle precipitation.

## 3 Results and discussion

### 3.1 Overall aerosol, cloud, and meteorological characteristics

To explore the effect of aerosols on cloud microphysics and the subsequent precipitation, a general understanding of aerosol quantities, cloud microphysics, and precipitation characteristics over the study region is crucial. Figure 3 illustrates the spatial distribution of mean aerosol and cloud parameter values (including AOD, COT, CWP, CF, CER, and CTP) over northern Taiwan from October 15 to November 30, 2005–2017. The mean AOD reached 0.6 in northwest Taiwan because of the high density of human activities, whereas lower AOD values (less than 0.2) were observed over the Xueshan Mountain Range (the green triangle in Fig. 3a).

Clouds were~~The characteristics of clouds are~~ affected by the prevailing northeast wind and topography, resulting in~~; therefore, clouds generally have~~ higher top heights and more significant coverage for clouds over northeastern Taiwan compared with northwestern Taiwan. The mean ~~COT,~~ CWP, CF and CER in our study area ~~had~~ ranged~~s of 10 g m$^{-2}$,~~ from 60–120 g m$^{-2}$, 0.6–0.7, and 13–14.5 µm, respectively. COT was usually around 10 and m~~The mean CF was 0.6–0.7. M~~ost of the CTP was higher than 850 hPa, suggesting low-level clouds (e.g., warm, thin, and broken clouds).~~For the spatial distribution of data availability, larger quantities were collected in our main research domain, indicating robust statistical results.~~

~~The characteristics of the s~~Surface $PM_{2.5}$ concentrations and meteorological parameters for clean and polluted days were also analyzed. We collected 1189 hours of rainfall data~~, and the number of other meteorological parameters data was~~ out of approximately 14,000 total hours of meteorological data. The mean values of temperature, relative humidity, rainfall, wind speed, and $PM_{2.5}$ concentrations were 22.3 °C, 74.9 %, 1.4 mm h$^{-1}$, 3.2 m s$^{-1}$, and 23.4 µg m$^{-3}$, respectively (illustrated in Fig. 4). The prevailing wind direction was northeast. During clean days, the aforementioned mean values were 22.2 °C, 79.3 %, 1.5 mm h$^{-1}$, 3.6 m s$^{-1}$, and 9.9 µg m$^{-3}$, respectively, compared with the mean values of 22.5 °C, 72.5 %, 1.4 mm h$^{-1}$, 2.7 m s$^{-1}$, and 43.3 µg m$^{-3}$, respectively, on polluted days. Overall, compared with clean days, meteorological conditions on polluted days had lower relative humidity, less rainfall, more wind direction in addition to the northeast wind, and lower wind speed. However, differences were not observed in mean rainfall rates between clean and polluted days. The number of rainfall hours differed significantly with 384 hours during clean days and 115 hours during polluted days. A weaker and more disorderly direction of the wind was observed on polluted days, which suggests that pollution may be associated with more stagnant conditions.

CWP is a constraint factor for the ACI index calculation illustrated in Eq. (1). We further examined CWP variability in response to main meteorological parameters (temperature, relative humidity, and rainfall) and $PM_{2.5}$ concentrations from the Pingzhen site and CER from MODIS. We calculated the daily mean value of CWP and CER by averaging grids over the main research area (24.6°–25.2° N, 120.9°–121.5° E). Daily meteorological parameters and $PM_{2.5}$ concentration data, described in Sect. 2.2, were used. Figure 5 illustrates the means and standard deviations of $PM_{2.5}$ and CER in 10 CWP groups. As CWP increased, the average temperature and relative humidity gradually decreased and increased, respectively. No significant correlation was identified between rainfall and CWP. The complicated relationship between $PM_{2.5}$ and CWP is illustrated in

Fig. 5. $PM_{2.5}$ increased with an increase in CWP to 50 g $m^{-2}$ and then decreased, whereas CER increased at first before decreasing and then increasing again. CWP standard deviation in group 9 (150 ≤ CWP < 297) was smaller than in other groups, indicating that group 9 was a more stable community; thus, much of the subsequent analysis focused on group 9 to reduce uncertainties caused by the variability of environmental conditions~~ and improve our understanding~~.

### 3.2 Aerosol effect on warm cloud properties

The effects of aerosols on warm cloud microphysics in different CWP groups for the main research domain were studied using the ACI index (Eq. (1)). Figure 6 illustrates the ACI values and correlation coefficient (r(ACI)) of the $PM_{2.5}$ mass concentration and CER under different CWP groups. ACI was 0.07 in CWP group 9 (150 ≤ CWP < 297) and had~~with~~ the lowest root mean square error (RMSE = 0.23) compared with other groups. The correlation coefficient between $PM_{2.5}$ and CER in group 9 was −0.19. Positive ACI values were observed when CWP was larger~~higher~~ than CWP group 7 (i.e. CWP groups 8-10)~~125 g m−2~~, and a higher value of ACI is associated with higher CWP groups. The negative correlation for these groups indicates an aerosol indirect effect (i.e. an increase in aerosols cause cloud droplet radii~~uses~~ to become smaller under a fixed water content). Negative ACI values were associated with~~are illustrated in~~ low CWP groups (i.e. groups 1–7), which may be caused by the large standard deviation of CER data in ~~smaller~~ CWP groups with lower values. However, in these low CWP groups~~the low water content~~ may reduce the effects of aerosols on warm cloud microphysics. We compared our results with values from the literature (Table). Feingold et al. (2003) analyzed ACIs by using ground-based remote sensors in Oklahoma, United States, focusing on ice-free, single-layered, nonprecipitating, and airborne insect–free clouds. Their results indicated that under the same LWP, the ACI values of seven cases were 0.02–0.16. Kim et al. (2008) conducted a three-year experiment by using ground-based remote sensors to investigate the aerosol indirect effect. Their results suggested that the ACI values of continental stratus clouds ranged from 0.04 to 0.17 in north-central Oklahoma. McComiskey et al. (2009) observed the ACI values of coastal stratiform clouds between 0.04 and 0.15 by using ground-based remote sensing data from the Atmospheric Radiation Measurement (ARM) program at Pt. Reyes, California, United States. Our~~Their~~ findings were on the lower end of these ranges, likely due to the more polluted conditions in our East Asia study area.~~indicated that values for the anthropogenic polluted area in the current study were on the lower end but within a reasonable range, despite an ACI value of 0.07 in the CWP group 9 (150 ≤ CWP < 297).~~

Because of the distinct ACI signal at CWP group 9, we further explored the effect of aerosols on cloud microphysical parameters by analysing their~~. The~~ differences ~~in cloud microphysics parameters~~ between polluted days and clean days over the main research area (24.6°–25.2° N, 120.9°–121.5° E)~~ was calculated~~. Compared with clean days, the COT, CER, CF, and CTT exhibited changes of +9.53, −2.77 µm, +0.07, and −1.28 K on polluted days (Fig. 7). While the positive CF value difference may have been due to higher aerosol loading, the atmospheric condition may have contributed as well. For instance, Saponaro et al. (2017) showed that CF is more sensitive to lower troposphere stability (LTS) than other cloud variables (i.e. CER, CTT, and COT). Also from Fig. 7, higher $PM_{2.5}$ concentrations corresponded to smaller CER and CTT values, and higher COT, in agreement with the aerosol indirect effect.~~These findings indicate that higher $PM_{2.5}$ concentrations may cause smaller~~

~~cloud droplet sizes under a high CWP environment, which accords with the concept of the aerosol indirect effect. Consequently, smaller droplets will reduce collision coalescence rates and suppress precipitation, thereby increasing the cloud lifetime, fraction, and optical depth.~~

The relationship between CTT and CER~~the cloud vertical profile~~ and aerosols was ~~also~~ studied in further detail. Figure 8 displays CWP group 9 (150 ≤ CWP < 297) results of the corresponding CTT-CER relationship and~~,~~ the occurrence frequency (%) ~~as a function of CTT and CER, and the vertical profiles of CER 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.~~highest occurrence frequency was located at a CER value of approximately 8 μm, which was similar on polluted days. However, CTT was lower on clean days, and the CER occurrence frequency increased overall. In other words, when the CTT was lower, the occurrence frequency of larger CER was higher.~~ This phenomenon could be caused by the onset of water cloud generation during strong~~when~~ updrafts, i.e. droplet size increases during air parcel expansion in an adiabatic process ~~dominate, causing adiabatic growth, and the reduction of in-cloud depth droplet size increases~~ (Saito et al., 2019). However, ~~O~~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~~the CER barely changed with a reduction in CTT. CTT between 285 and 288 K~~ exhibited a higher occurrence frequency ~~during polluted days, whereas clean days had a higher frequency of CTT~~ between 288~~282~~ 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. ~~high aerosol concentrations introduced higher concentrations of CCN, produced more liquid particles at warmer CTT, and inhibited the development of cloud droplet size.~~

### 3.3 Effect of different polluted conditions on ACI

We further explored the effect of aerosols on cloud microphysics under different polluted conditions. We investigated ACI from two perspectives, considering different polluted levels and considering different polluted areas. First, we divided polluted days into three equal groups: slightly, moderately, and heavily polluted days. We then calculated ACI values by using RMSE and correlation coefficients (denoted with r(ACI)) of $PM_{2.5}$ and CER under different CWP groups and at different polluted levels for the main research domain. As illustrated in Fig. 9a, three polluted levels exhibited similar trends, but stronger ACI signals (larger ACI slope and absolute r(ACI) values) were observed for heavily polluted cases compared with moderately and slightly polluted days. On heavily polluted days (red line), when the CWP was larger~~higher~~ than group 5~~50 g m$^{-2}$~~, the ACI value increased with increasing~~as~~ CWP, and from group 8 ~~increases. When CWP increased to group 8~~, the ACI value was positive ~~for polluted days~~, whereas ACI values for slightly and moderately polluted days continued to increase in groups 7 to 9 but decreased in group 10 and were not consistently at positive ACI values past a particular CWP range. For CWP ~~in~~ groups 7–10, the ACI values of heavily polluted days were consistently higher than the ACI values of slightly and moderately polluted days, especially in group 10. Notably, the differences in ACI values for the three polluted levels (0.08, 0.07, and 0.06 for heavily, moderately, and slightly, respectively) associated with CWP group 9 (150 ≤ CWP < 297) were apparently small, thus the effects on cloud properties may prove insignificant. ~~CWP at group 9 ($150 \le$ CWP $< 297$) displayed consensus on ACI and r(ACI) values, indicating that clouds with a CWP range of 150–297 g $m^{-2}$ were sensitive to aerosol indirect effects. Under high pollution, aerosols had a larger effect on cloud microphysics and larger positive ACI values (0.08, 0.07, and 0.06 for heavily, moderately, and slightly, respectively).~~

The effects of aerosols on cloud microphysics over the land and ocean (denoted with magenta and blue square boxes, respectively, in Fig. 3) are discussed. Because of the lack of $PM_{2.5}$ surface observations ~~of $PM_{2.5}$~~ over the ocean, we used AOD from MODIS/Aqua as the aerosol proxy in Eq. (1) for the ACI calculation. To ensure the reliability of calculations, we computed ACI in the primary research area (24.6°–25.2° N, 120.9°–121.5° E) based on different aerosol proxies (i.e. AOD and $PM_{2.5}$ concentration). As illustrated in Fig. 9b, in CWP groups 1–8, ACI values evaluated with AOD had larger values than those evaluated with $PM_{2.5}$; the difference was the largest in CWP group 2 (0.22). For positive ACI ranges, ACIs estimated with AOD were positive for CWP groups 7–10, whereas ACIs computed with $PM_{2.5}$ were positive after CWP group 8. In CWP groups 8–10, differences in ACI values became smaller, especially in group 9. We focused on group 9, which had an ACI value using $PM_{2.5}$ of 0.07 and an ACI value using AOD of 0.06, a~~; the~~ difference of ~~between the two calculations is~~ only 0.01.

The effects of aerosols on cloud microphysics in polluted (i.e. land) and remote (i.e. ocean, mean AOD of 0.31) areas can be assessed further by using the ACI value with AOD as an aerosol proxy. We defined the main research area of 24.6°–25.2° N and 120.9°–121.5° E as the polluted area (Fig. 3a magenta box) and 25.2°–5.8° N and 120.9°–121.5° E as the remote area (Fig. 3a blue box). As illustrated in Fig. 9c, ACI values and correlation coefficients between mean AOD and CER were calculated in remote and polluted areas. Comparing ACI values between polluted and remote areas demonstrated that ACI values were higher in the polluted area in CWP groups 1–5. In this CWP interval, the ACI values of the remote area increased with an increase in CWP, whereas the ACI values of the polluted area changed significantly. In CWP groups 6–10, the ACI values of the remote area became more pronounced than the polluted area. The positive and increasing tendency of ACI values was observed in larger CWP groups ($>7$) in two areas, suggesting that the environmental condition (i.e. water vapor) was critical in aerosol indirect effects. In CWP group 9, ACI values were 0.09 and 0.06 for remote and polluted areas, respectively, indicating that aerosol indirect effects were~~are~~ stronger in remote areas (i.e. lower aerosols). These results are consistent with a study (Saponaro et al., 2017) that found~~reported that~~ large aerosol concentrations can saturate the effect of ACI causing a lower ACI value.

### 3.4 Aerosol effect on precipitation

Aerosol effects on warm cloud properties were discussed in Sect. 3.2; these effects may subsequently alter the cloud lifetime and the precipitation process. This section further explores their consequential influence on precipitation.~~The presence of aerosols enhances the concentration of condensation nuclei under the quantitative water content, which increases the cloud droplet number, reduces CER, and increases COT and CF. These changes subsequently alter the cloud lifetime and the precipitation process. This section further explores their consequential influence on precipitation.~~ High-time resolution (one-minute) JWD and $PM_{2.5}$ datasets were used to investigate the effects of aerosols on the raindrop size distribution, rainfall, and cloud lifetime. 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.

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), 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.

~~The number of sample occurrences under different raindrop size classifications for clean and polluted days (not displayed here) were significantly higher on clean days, suggesting that a cleaner environment could be favourable for raining, compared with polluted environments. We further calculated the daily-averaged droplet number in each raindrop size classification for polluted and clean days. The difference is plotted as Fig. 10a. The results illustrate that during polluted days, the droplet numbers appear lower for the smaller raindrop bins (≤ n8) and higher for the larger raindrop bins (> n8). A significant reduction~~

~~in droplet numbers (decreased from 68 in clean days to 56 in polluted days) was observed at the n2 bin, which represents the reduction in drizzle. Our preliminary findings suggest that cloud condensation nuclei may have competing effects on water uptake under aerosol-laden air and cloud water content limited conditions, which would alter the precipitation processes.~~

~~To investigate the effect of the high aerosol concentration on light rain, we created a similar plot to those that we had previously constructed but only for precipitation of less than or equal to 1 mm h$^{-1}$ (Fig. 10b). 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, indicating that drizzle drops were a common raindrop type when rainfall was ≤1 mm h$^{-1}$. Instead of the daily average number used in Fig. 10a, 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. The difference between polluted and clean days is illustrated in Fig. 10b. 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 2.35 % more), whereas polluted days accounted for a significantly lower proportion of n3 to n8 (Fig. 10b). These results indicate that if precipitation is lower than or equal to 1 mm h$^{-1}$, a high aerosol concentration drives raindrops to move towards smaller drop sizes, which increases the appearance of drizzle drops.~~

A modeling study (Huang et al., 2007) revealed that the second ~~aerosol~~ indirect effect of aerosols (a large number of small droplets are generated by~~and~~ enhanced aerosols and reduce the ~~cloud~~ precipitation efficiency) significantly reduces fall and winter precipitation from 3 % to 20 % across East Asia, although it was dependent on the auto-conversion scheme assumed. In this study, we used observational data (i.e. JWD) to analyze the difference between the average daily rainfall of polluted and clean days in different CWP groups and explored whether the increase in aerosol loading inhibits precipitation. Figure 12a~~Figure 11a~~ demonstrates that the daily rainfall difference between polluted and clean days varies greatly~~largely~~ between CWP groups 1–7, which may be ~~because of the~~ small~~er~~ sample numbers in the those CWP groups~~interval~~. However, the average daily rainfall on clean days consistently exhibited~~s~~ higher values in CWP groups 8–10 compared with polluted days. In CWP group 9 (150 ≤ CWP < 297), the daily average rainfall on polluted days (1.4 mm) decreased by 6.8 mm compared with clean days (8.2 mm). ~~Our findings suggest that under the fixed cloud water content, precipitation tends to decrease in high aerosol loading environments, which echoes findings reported in Sect. 3.2.~~

Furthermore, we analyzed the hourly rainfall rate of CWP group 9 (150 ≤ CWP < 297) for clean and polluted days to explore the effect of aerosol on cloud lifetime. ~~(i.e. the cloud lifetime effect (Albrecht, 1989; Pincus and Baker, 1994; Lohmann and Feichter, 2005)). Figure 11b~~Figure 12b illustrates the ~~24-hour~~ rainfall rate trends for clean and polluted days. On clean days, rainfall was~~is~~ randomly distributed throughout the entire day with a notably larger rainfall rate observed after 04:00~~4 am~~, whereas no rainfall was observed during daytime on polluted days, and a relatively weak rainfall rate started early in the night. Although the existence of an aerosol effect on cloud lifetime is still widely disputed (Small et al., 2009; Stocker, 2014), ~~O~~our preliminary results show~~suggest~~ that ~~aerosols can suppress and delay~~ precipitation might be suppressed and delayed under high aerosol loading. Combined with the results from Sect. 3.2, the process in the aerosol-cloud-precipitation interactions is consistent with the cloud lifetime effect. The presence of aerosols enhances the concentration of condensation nuclei under a fixed water content, which increases the cloud droplet number, redistributes cloud water to more numerous and smaller droplets, reducing collision–coalescence rates, which in turn suppresses precipitation and delays rainfall occurrence (i.e. the cloud lifetime effect (Albrecht, 1989; Pincus and Baker, 1994; Lohmann and Feichter, 2005)).~~. Under a constant liquid water content, a higher aerosol concentration redistributes cloud water to more numerous and smaller droplets, reducing collision–coalescence rates, which in turn suppress precipitation and delays rainfall occurrence.~~ Our results provide evidence of this and other aerosol indirect effects over~~in~~ a highly populated island in the western Pacific.

## 4 Conclusions

Numerous studies have explored aerosol-cloud-precipitation interactions in marine stratocumulus clouds based on in-situ observations, satellite observations, and models; however, few studies have investigated clouds over a dense population and complex topography area. In this study, we integrated numerous aerosol, cloud, and precipitation data from satellite and surface observations to quantify the effects of aerosols on low-level warm cloud microphysics and precipitation over northern Taiwan, an urban area on the northwest Pacific Ocean. A 13-year (2005–2017) dataset with a selected time frame (October 15 to November 30) was used in this study. In contrast to previous studies that have focused on the rainfall rate, we investigated changes in raindrop size distribution as the key variable in the effect of aerosols on precipitation.

We used surface $PM_{2.5}$ mass concentration data as aerosol proxy to study~~determine~~ the aerosol impacts on clouds and precipitation~~indirect effect~~. According to~~Based on the~~ $PM_{2.5}$ mass concentration level, the data ws~~we~~ split ~~the observations~~ into clean and~~,~~ polluted days.~~, slightly polluted, moderately polluted, and heavily polluted groups.~~ The analysis of aerosol effects on clouds indicated that in CWP group 9 ($150 \leq CWP < 297$), the average~~ of~~ 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~~in~~ 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.~~Results illustrate that increasing the aerosol loading increases the cloud droplet number concentration and reduces the cloud droplet size under fixed water content, thereby increasing the cloud lifetime, increasing the CF, and allowing clouds to develop further; these results are consistent with the aerosol indirect effect. The~~ 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. ~~of warm clouds in clean and polluted days indicates that higher aerosol concentrations produced more liquid particles at lower altitude and inhibited the development of the cloud droplet size under the air-polluted condition.~~ 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 ~~revealed that the~~ 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 ~~By~~ combined~~ing~~ 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.~~, we determined that the frequency of drizzle in the polluted conditions was decreased, whereas the high aerosol concentration caused a reduction in raindrop sizes, which increased the appearance of drizzle drops in the scenario of low precipitation (≤1 mm $h^{-1}$).~~

Our observational results from northern Taiwan in fall show~~ in~~ agreement with the aerosol indirect effects. However, we did not consider the aerosol direct radiative effect~~ and~~ or long-term variations caused by different weather systems~~ in the long-term statistic~~. Overall, this study used long-term surface and satellite data for a preliminary understanding of aerosol variations in northern Taiwan, the effects of aerosol on the environment, and the effects of aerosols on the precipitation. We suggest that further research~~es~~ on aerosol–cloud–precipitation interactions over this area should~~need to~~ be conducted~~carried out~~ to fully understand these processes.

*Data availability*. The satellite data from the MODIS instrument used in this study were obtained from https://ladsweb.modaps.eosdis.nasa.gov/search/. The meteorological and $PM_{2.5}$ observation data were available from Taiwan EPA at https://erdb.epa.gov.tw/FileDownload/FileDownload.aspx. JWD disdrometer data in this study were provided by the Planetary Boundary Layer and Air Pollution Lab. of the Dept. of Atmospheric Sciences, National Central University of Taiwan (http://pblap.atm.ncu.edu.tw/weather10.asp).

*Competing interests*. The authors declare that they have no conflict of interest.

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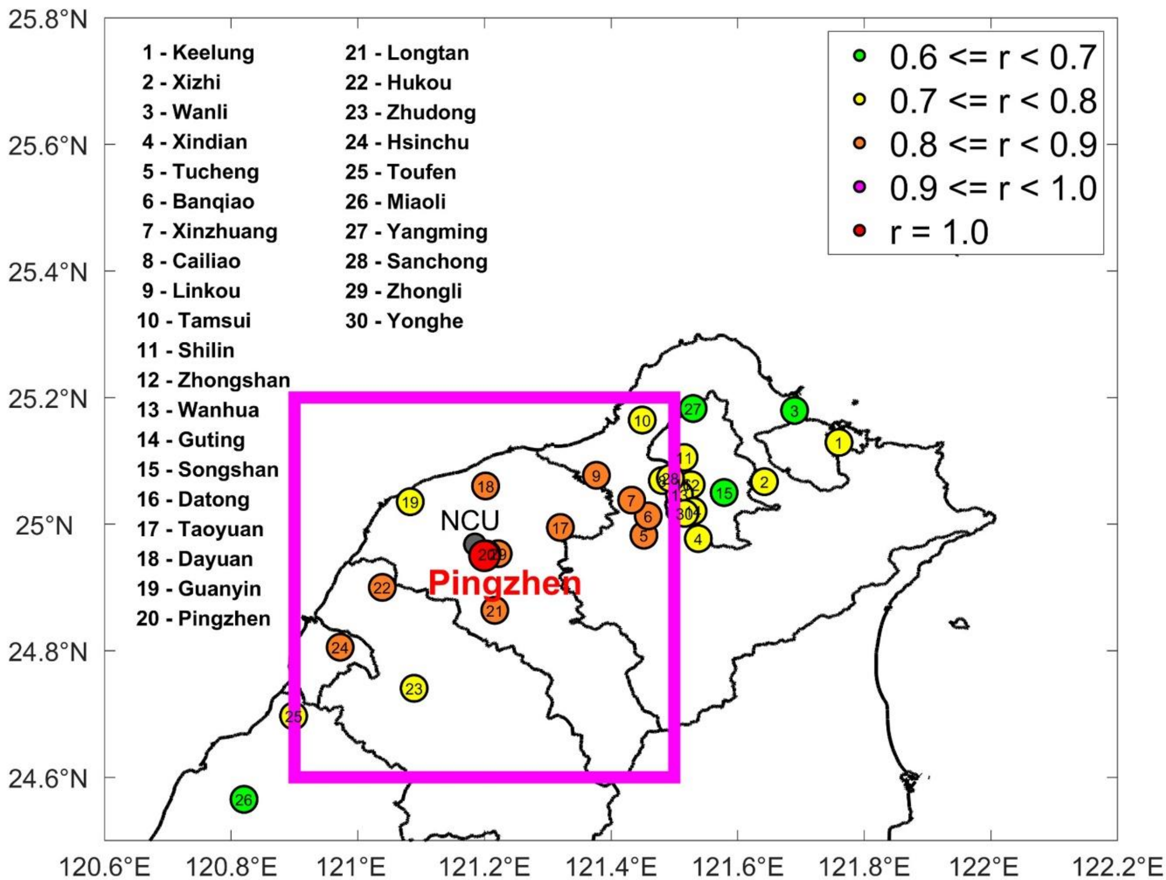
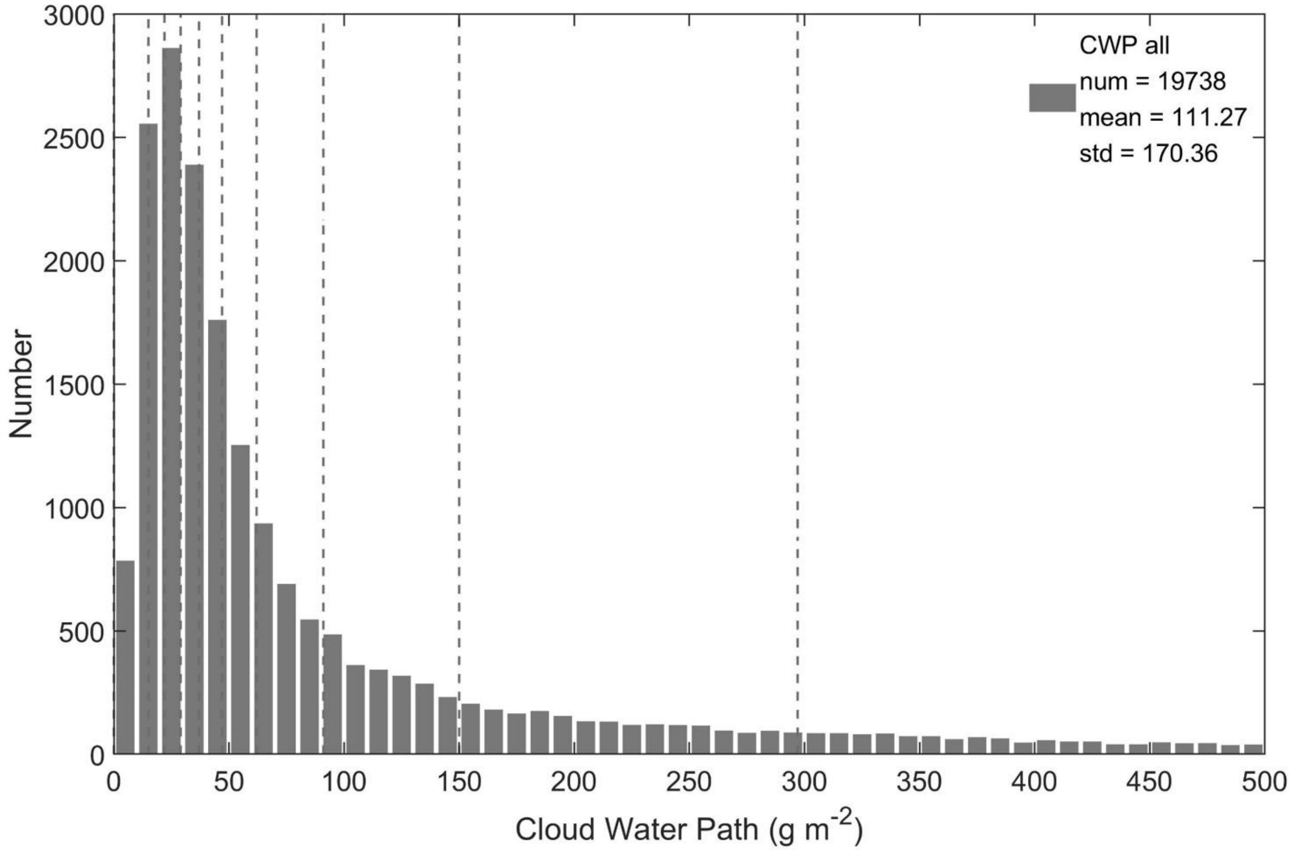



**Figure 1: Spatial correlation coefficient of the $PM_{2.5}$ concentration between Pingzhen station and other stations. The main research area (24.6°–25.2° N, 120.9°–121.5° E) is indicated with a magenta box.**


3000
2500
2000
1500
1000
500
0
Number
0
50
100
150
200
250
300
350
400
450
500
Cloud Water Path (g m-2)
CWP all
num = 19738
mean = 111.27
std = 170.36

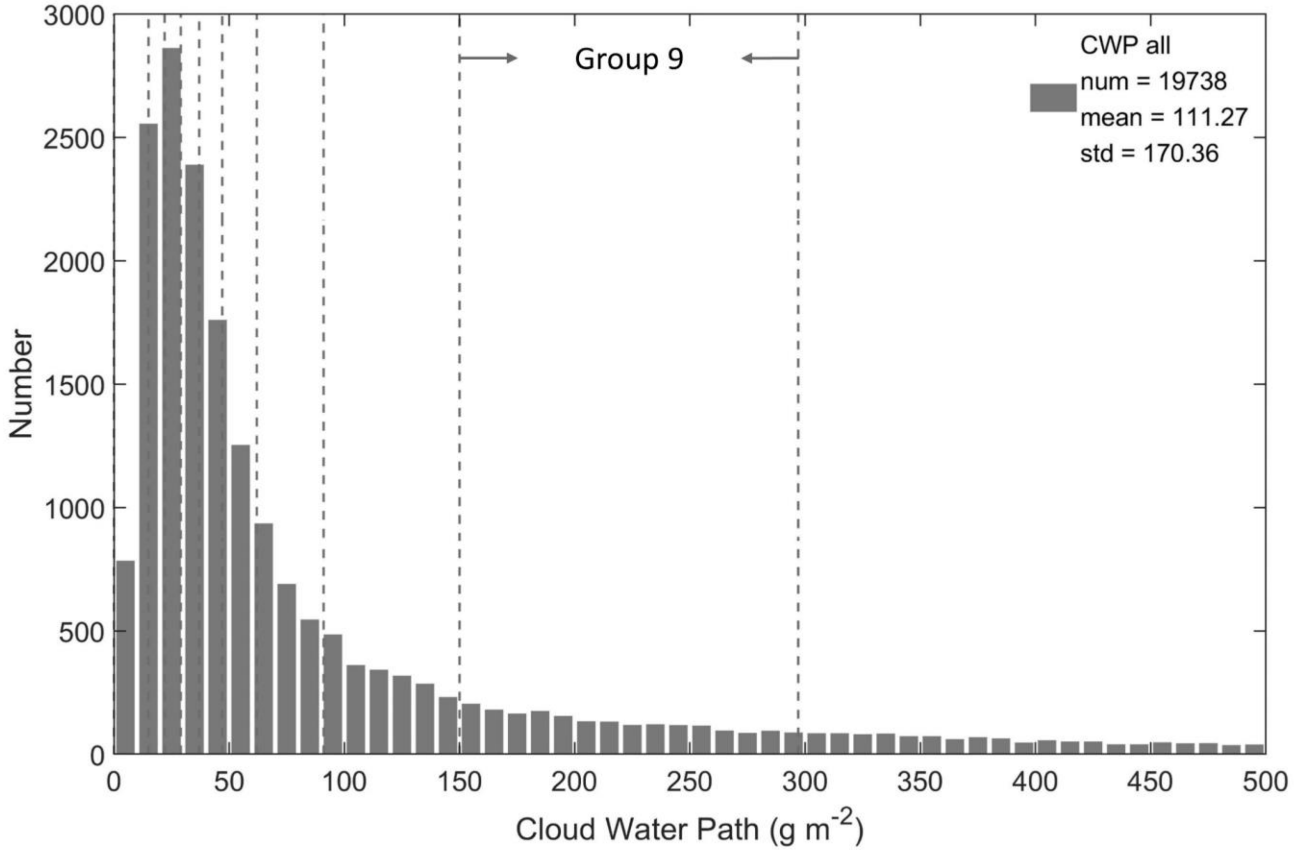



**Figure 2: Histogram of cloud water path (CWP) values over northern Taiwan from Oct. 15 to Nov. 30, 2005–2017. The CWP is divided into 10 bins (10 % for each bin) indicated by dashed lines. The key CWP group 9 (150 ≤ CWP < 297) is marked in the figure.**

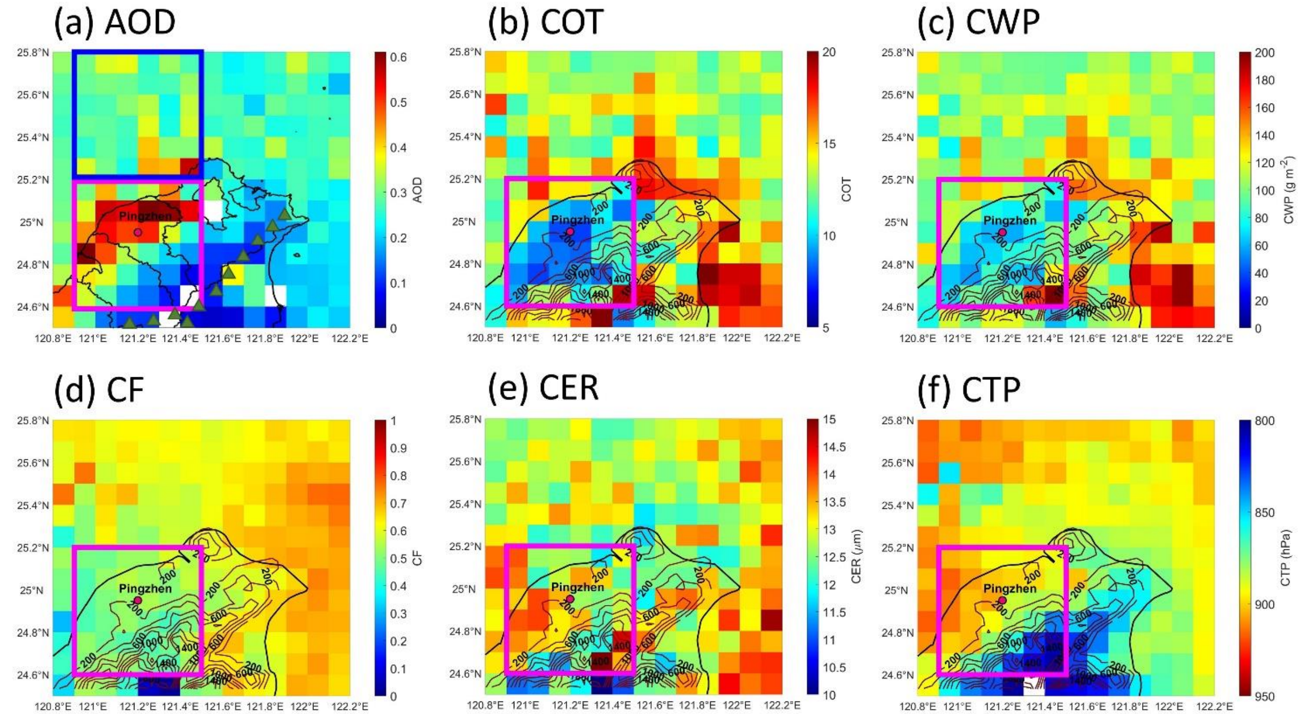



**Figure 3: Average (a) aerosol optical depth (AOD), (b) cloud optical thickness (COT), (c) cloud water path (CWP), (d) cloud fraction (CF), (e) cloud effective radius (CER), and (f) cloud top pressure (CTP) in warm clouds from October 15 to November 30, 2005–2017. The magenta box represents the main study area (24.6°–25.2° N, 120.9°–121.5° E) and the blue box in (a) is the remote area (25.2°–25.8° N, 120.9°–121.5° E). The green triangles in (a) mark the location ~~represent the schematic~~ of the Xueshan Mountain Range. The topography of north Taiwan is depicted with brown color contour lines (in meter) in (b)–(f).**

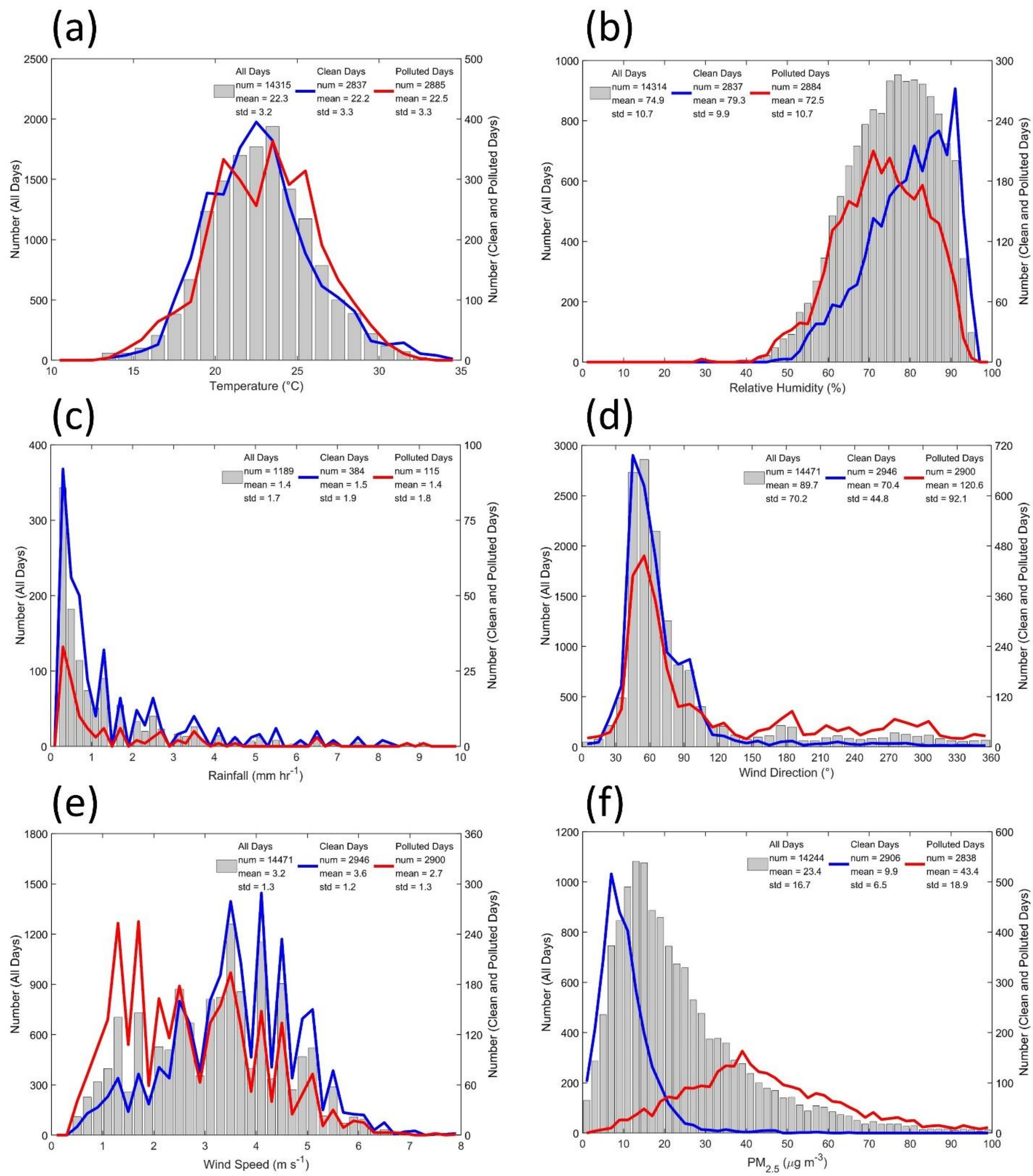
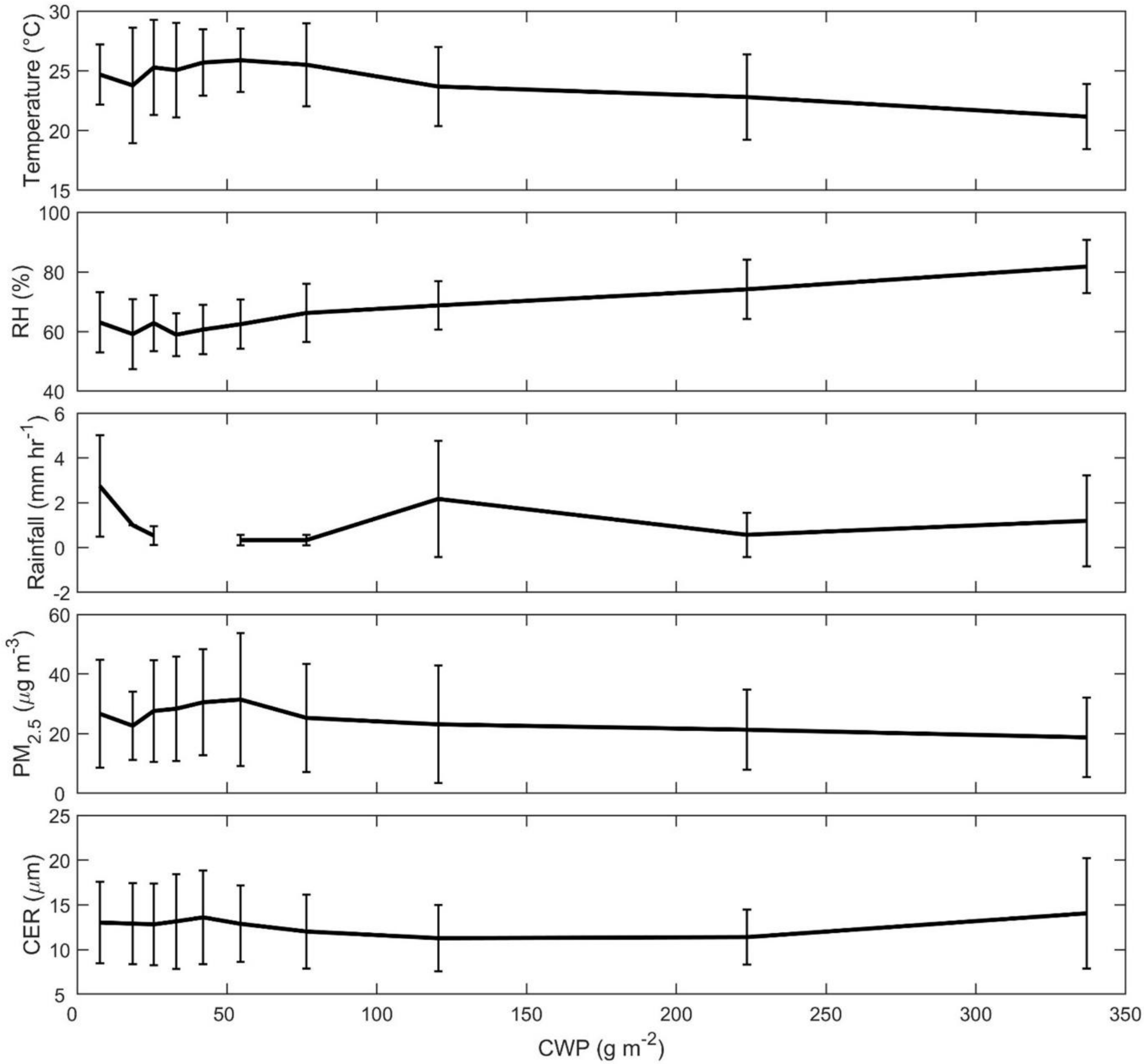



**Figure 4: The distribution of (a) temperature, (b) relative humidity, (c) rainfall, (d) wind direction, (e) wind speed, and (f) $PM_{2.5}$ hourly data from Pingzhen station from Oct. 15 to Nov. 30, 2005–2017. The gray bars are the distribution of all valid observations, the blue lines represent the clean days and the red lines represent the polluted days.**



Temperature (°C)
RH (%)
Rainfall (mm hr$^{-1}$)
PM$_{2.5}$ ($\mu$g m$^{-3}$)
CER ($\mu$m)
CWP (g m$^{-2}$)

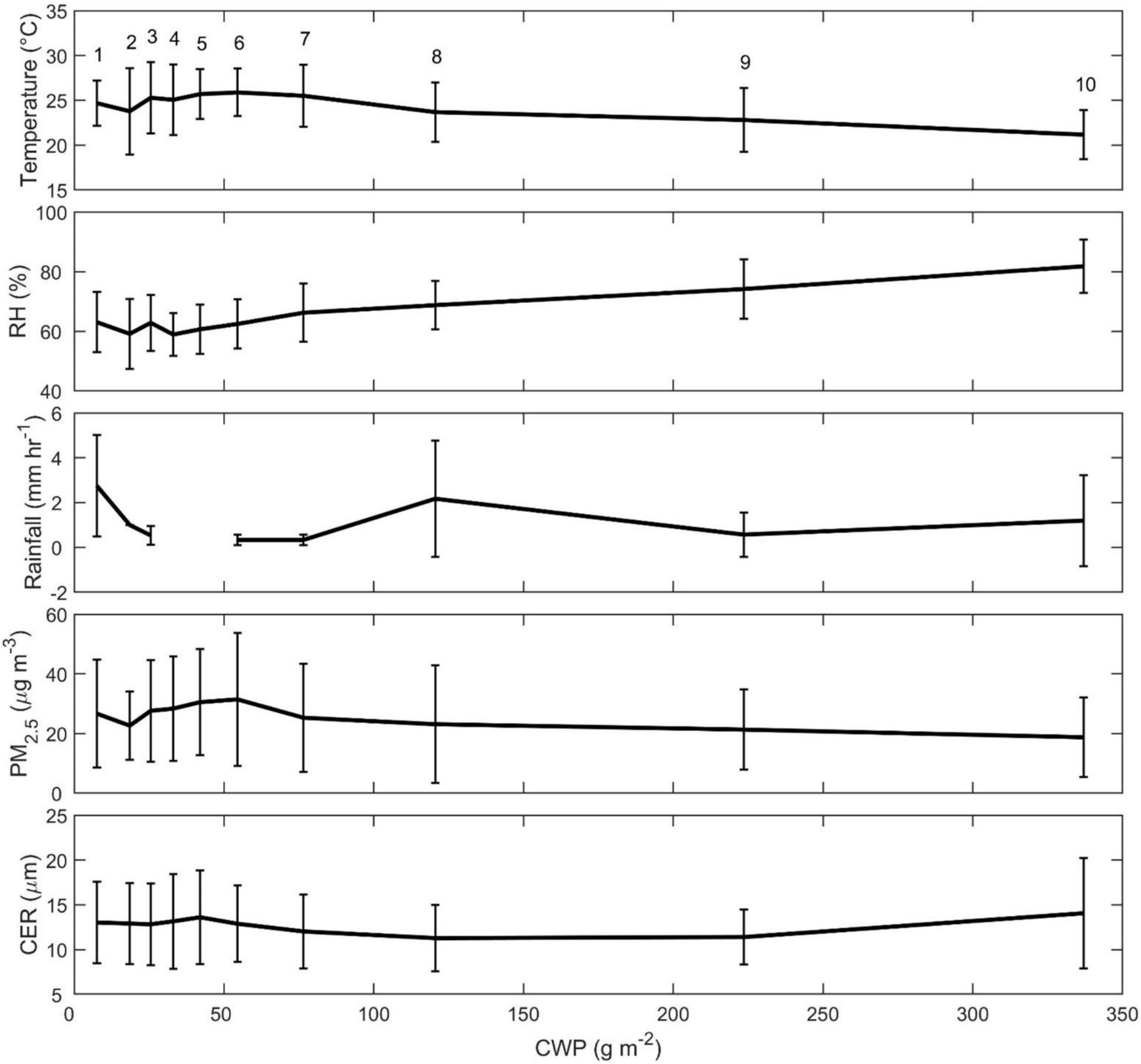
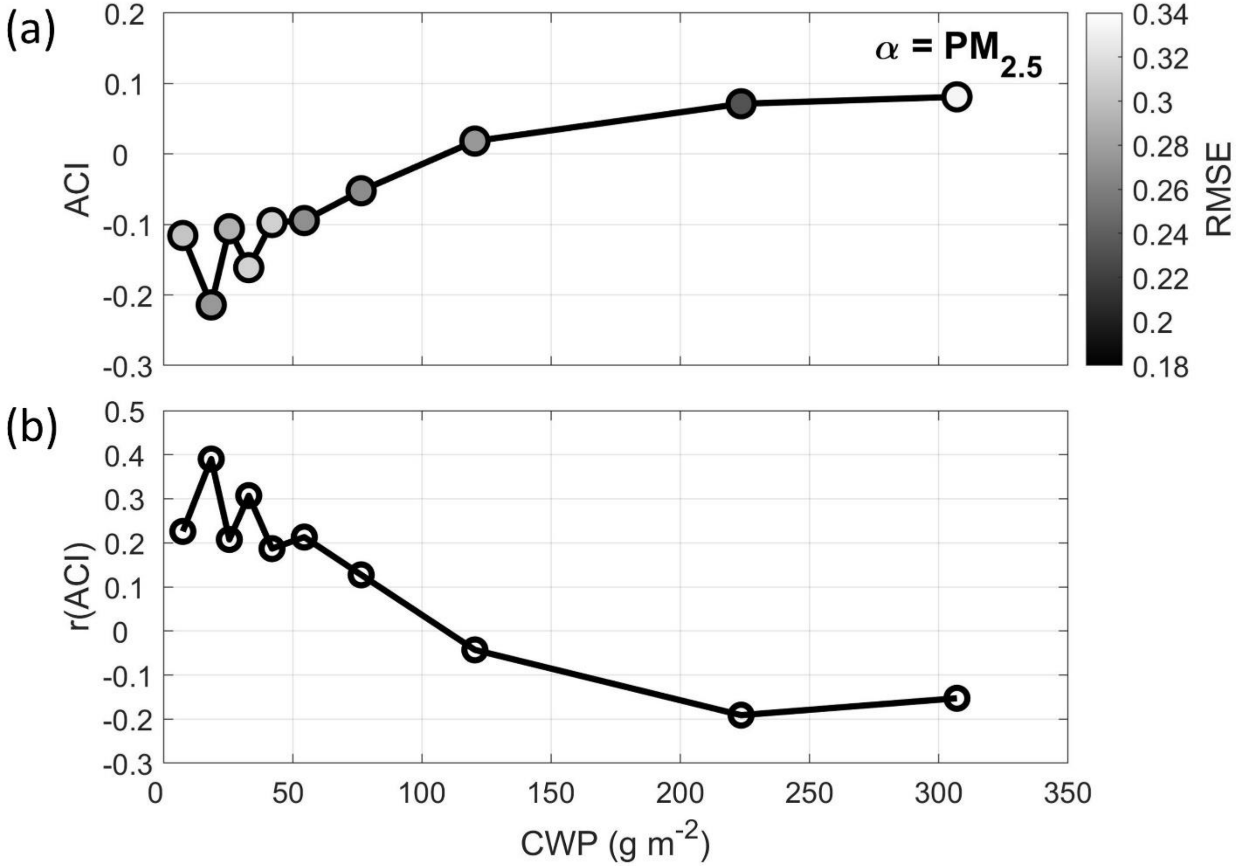



**Figure 5: Multiyear (2005–2017) mean and standard deviation of temperature, relative humidity (RH), rainfall, PM$_{2.5}$, and cloud effective radius (CER) in different cloud water path (CWP) bins.** **The CWP group numbers are marked in the top panel.**


(a)
α = PM2.5
ACI
RMSE
0.34
0.32
0.3
0.28
0.26
0.24
0.22
0.2
0.18
(b)
r(ACI)
CWP (g m-2)
0
50
100
150
200
250
300
350

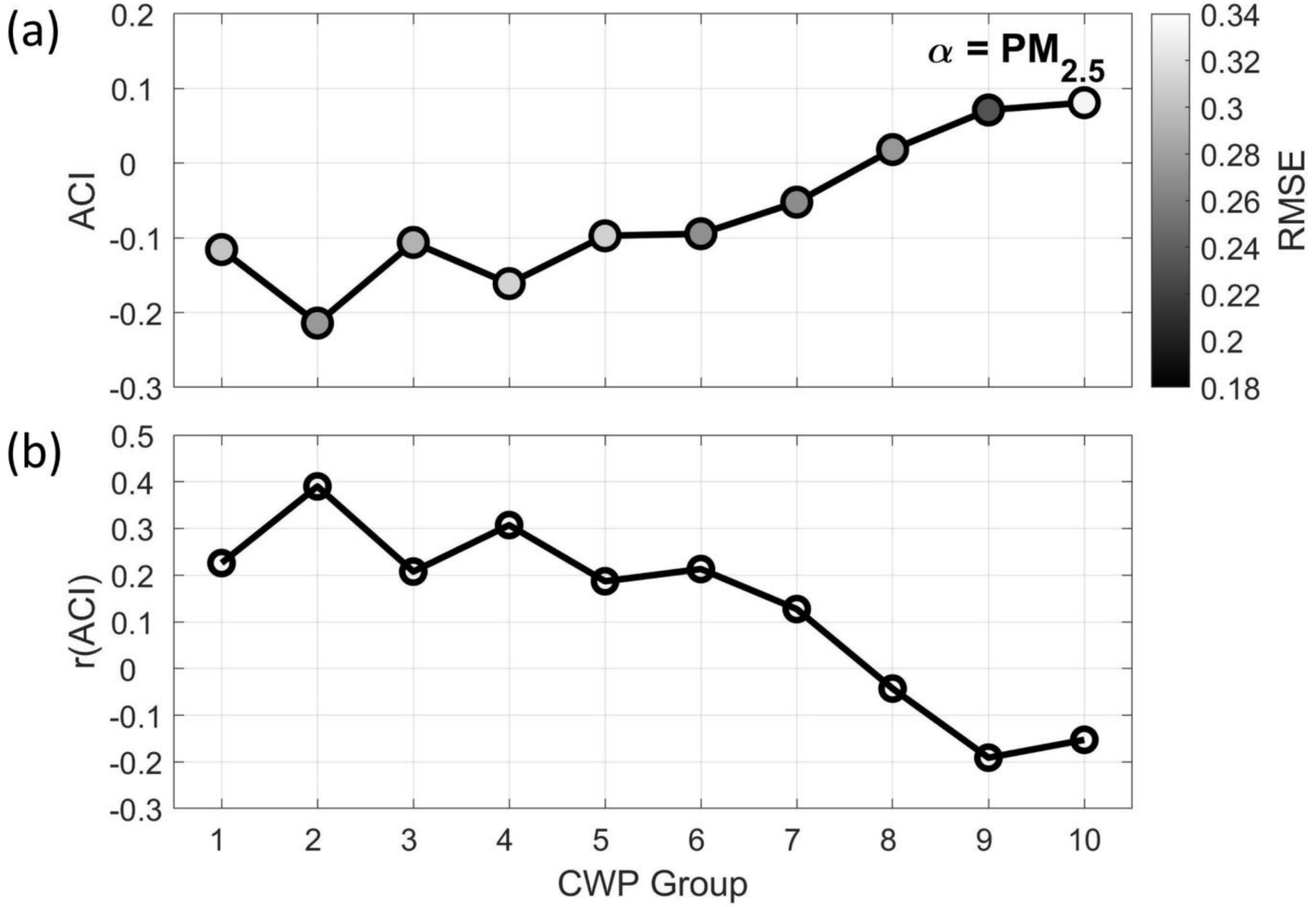



**Figure 6: (a) Aerosol cloud interaction (ACI) estimated values, computed for the cloud effective radius (CER) in the different CWP groups by applying the $PM_{2.5}$ concentrations as aerosol proxies. The shading in (a) represents the RMSE. (b) The correlation coefficients between $PM_{2.5}$ and CER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

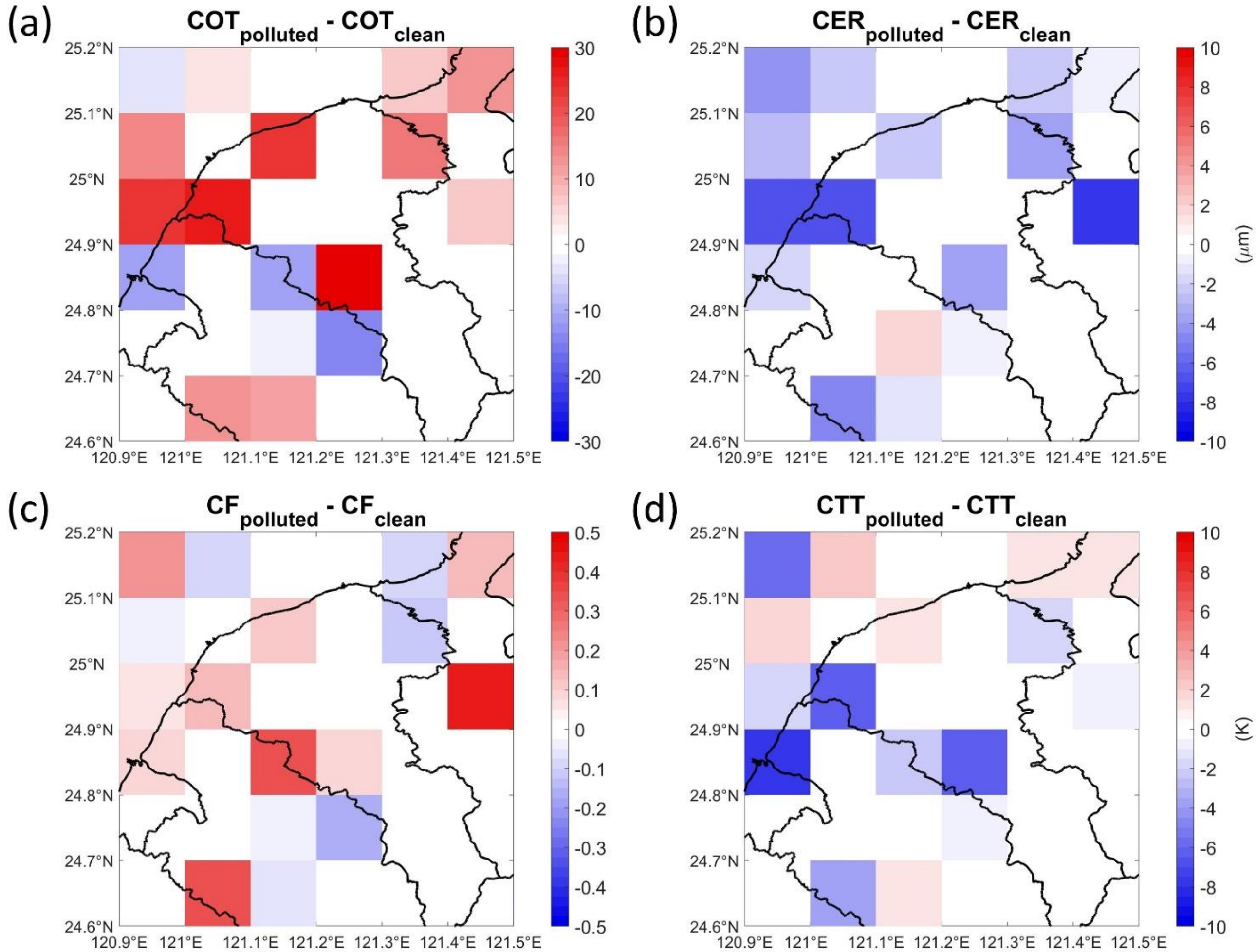
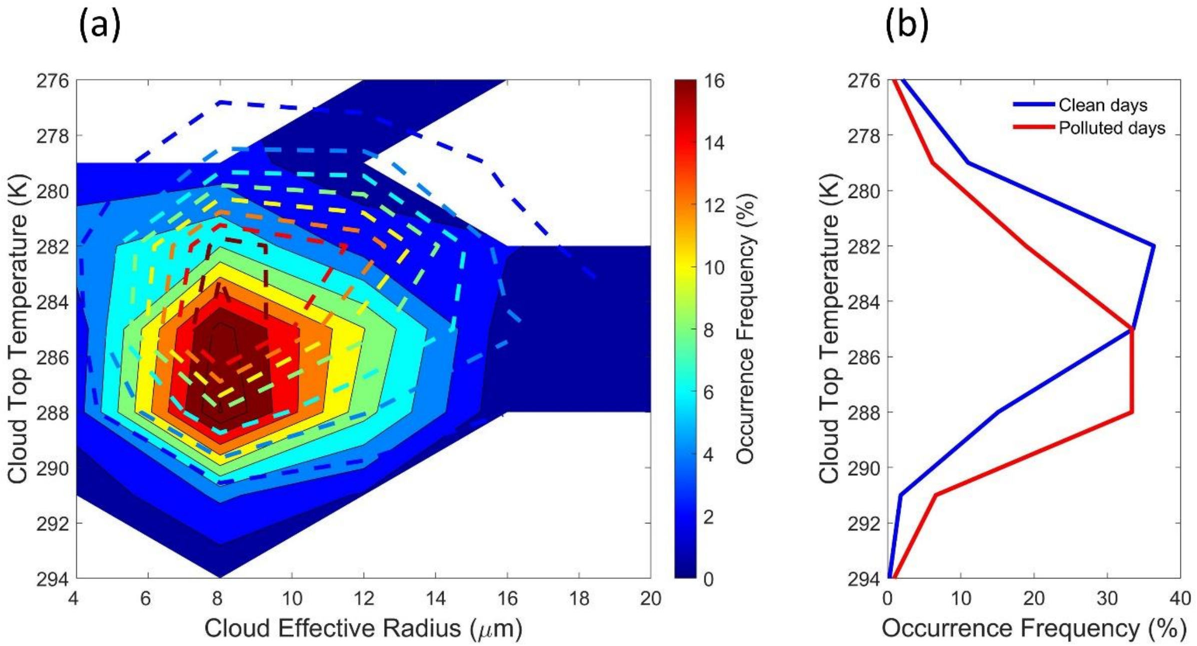



**Figure 7: Difference in (a) COT, (b) CER, (c) CF, and (d) CTT between polluted days and clean days in group 9 (150 ≤ CWP < 297).**



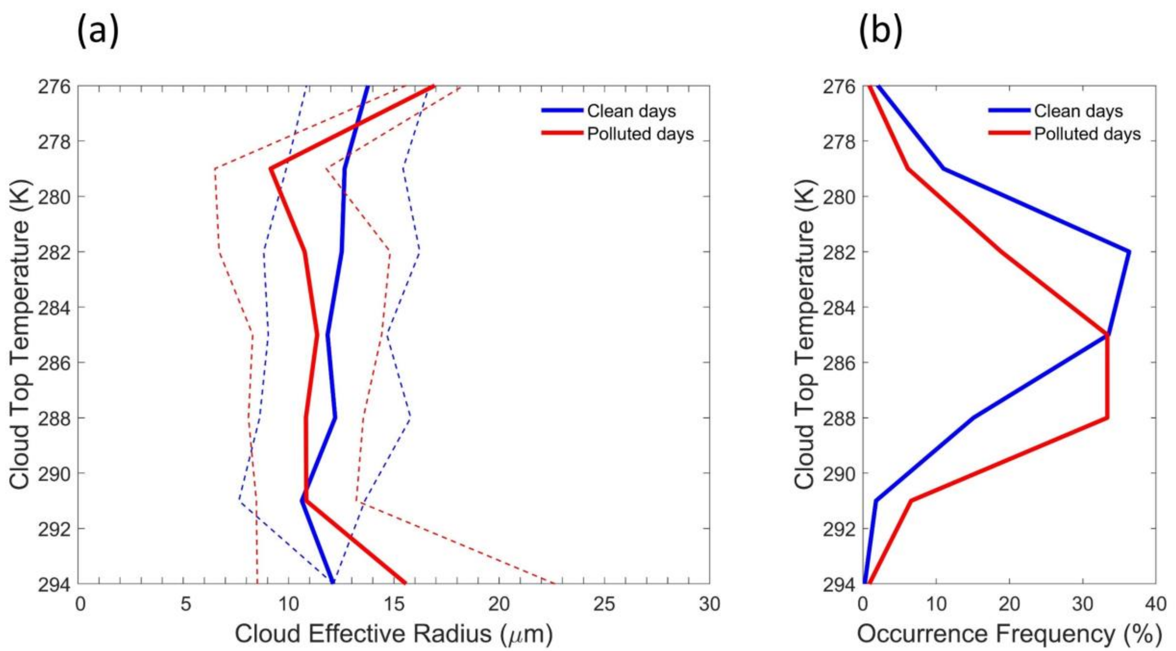
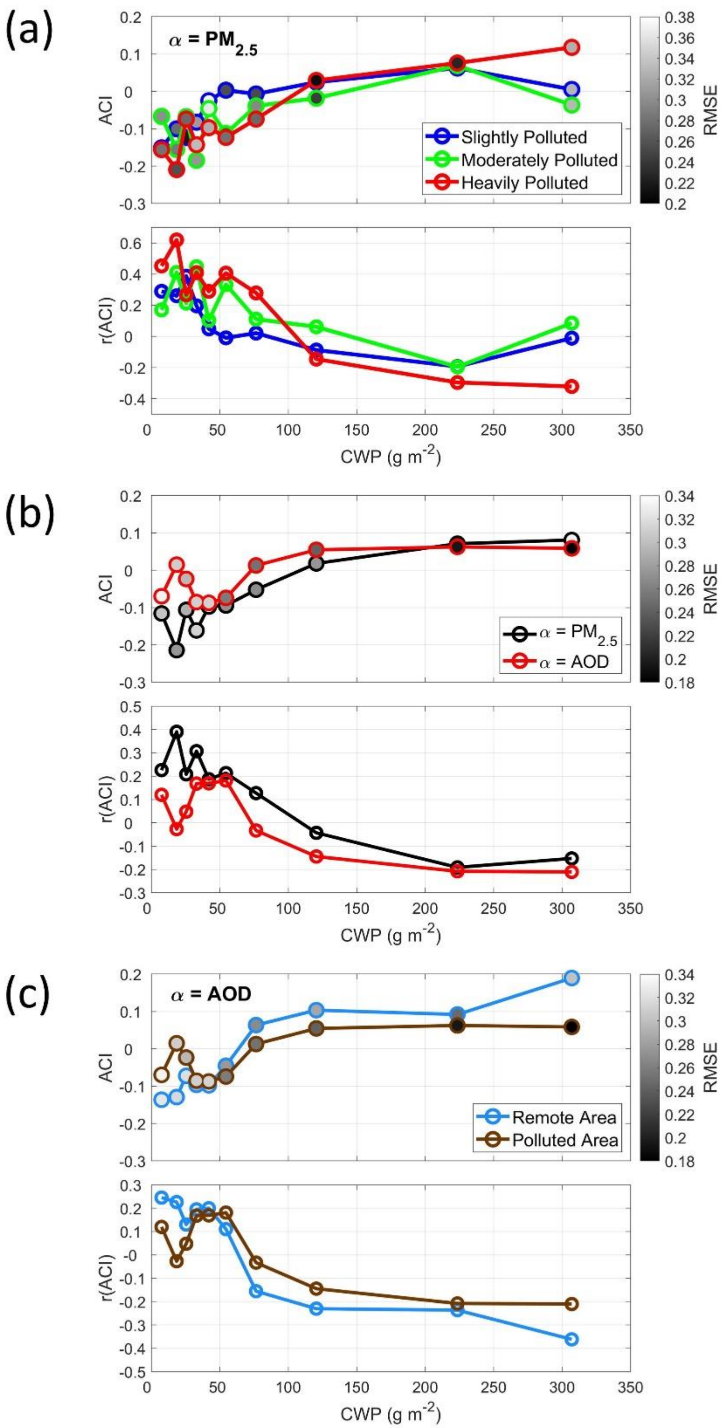




**Figure 8: Multiyear (2005–2017) (a) ~~occurrence frequency (%) as a function of~~ cloud top temperature (CTT)~~- and~~ cloud effective radius (CER) relationship. Plotted are the mean (solid line) and one standard deviation (dashed line) of the CER for each 3 K interval, and ~~The color contour in the shaded area and the dashed lines denote the polluted and clean days, respectively.~~ (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 ≤ CWP < 297).~~The vertical profile of CTT occurrence frequencies for polluted and clean days are depicted with red and blue lines, respectively. Both in CWP group 9 (150 ≤ CWP < 297).~~



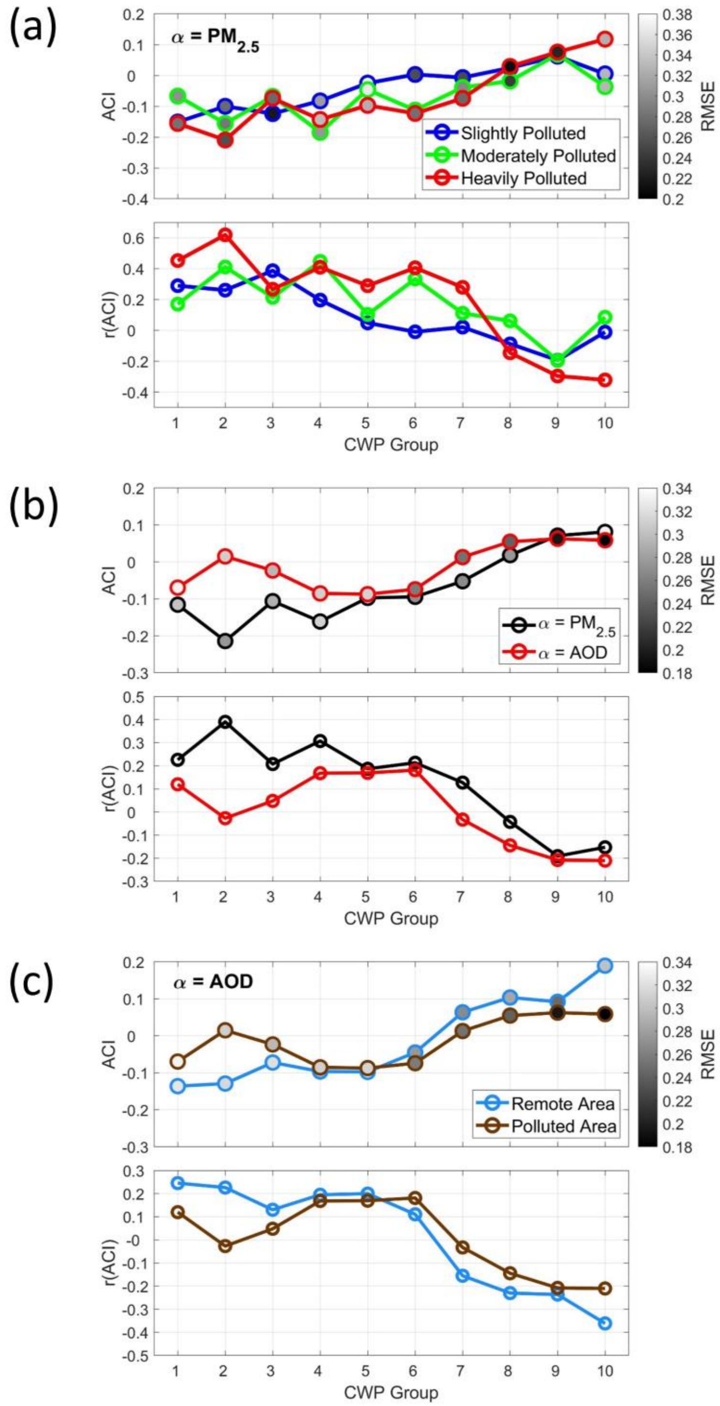
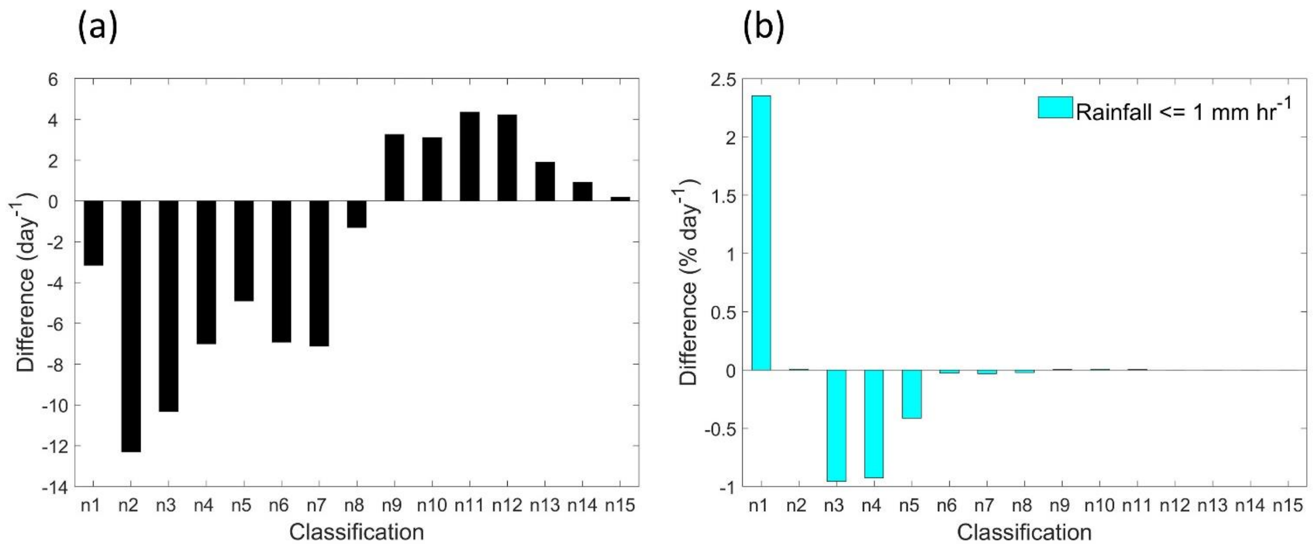



Figure 9: Multiyear (2005–2017) ACI values with the RMSE (shaded) and the correlation coefficient among (a) different polluted levels, (b) different aerosol proxies, and (c) different polluted condition areas.



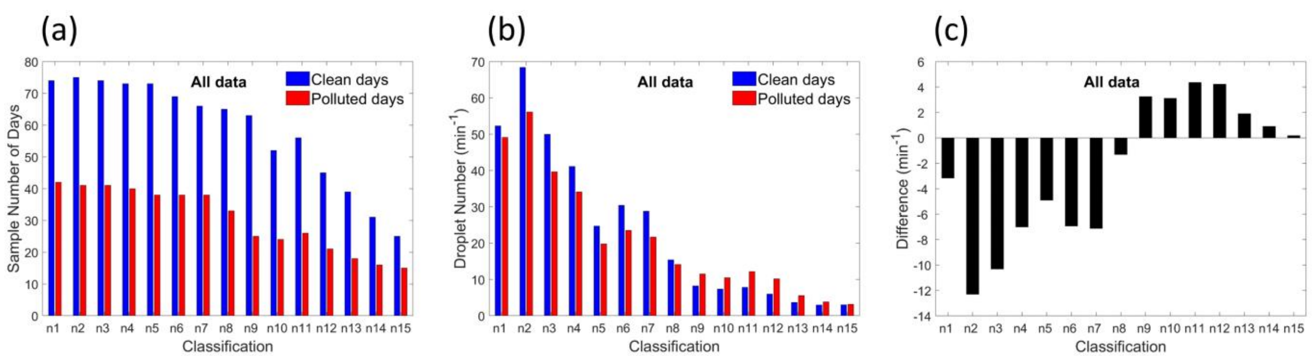




**Figure 10: Multiyear (2005–2017) (a) JWD sample number of days in each raindrop size bin, (b) mean droplet number per minute for clean and polluted days and (c) The differences in the mean droplet number between polluted and clean days. ~~(a) differences in the mean droplet number between polluted and clean days and (b) differences between polluted and clean days in the percentage of the cumulative droplet number distribution in different precipitation scenarios less than or equal to 1 mm h−1.~~ nX reflects different raindrop size bins. The 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.**

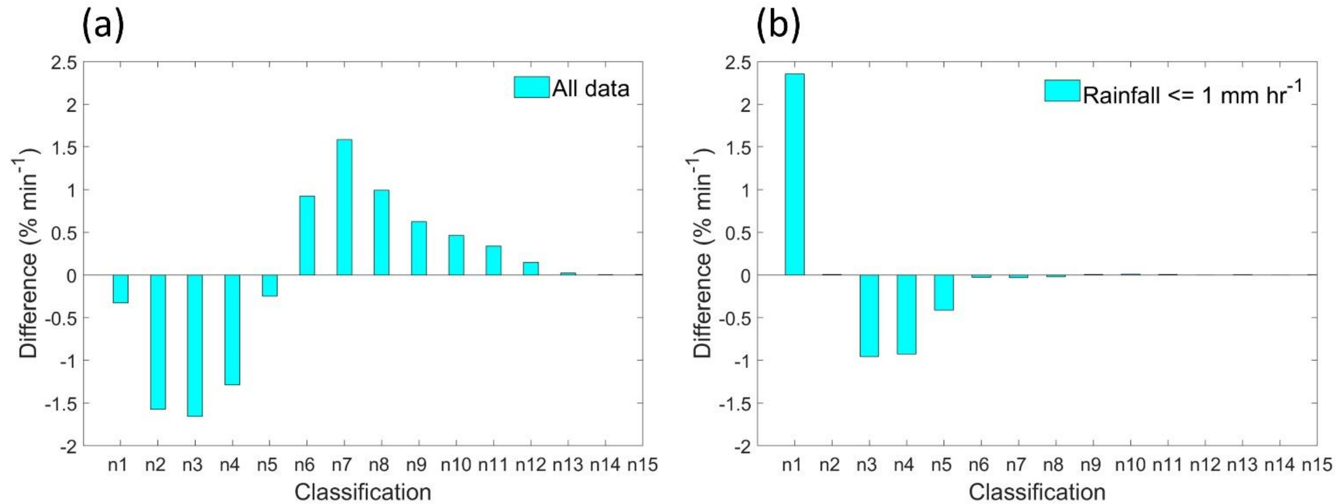



**Figure 11: Multiyear (2005-2017) differences between polluted and clean days as percentages of the cumulative droplet number distribution for (a) all data and (b) the data with precipitation less than or equal to 1 mm h$^{-1}$. nX reflects different raindrop size bins as listed in Fig. 10.**

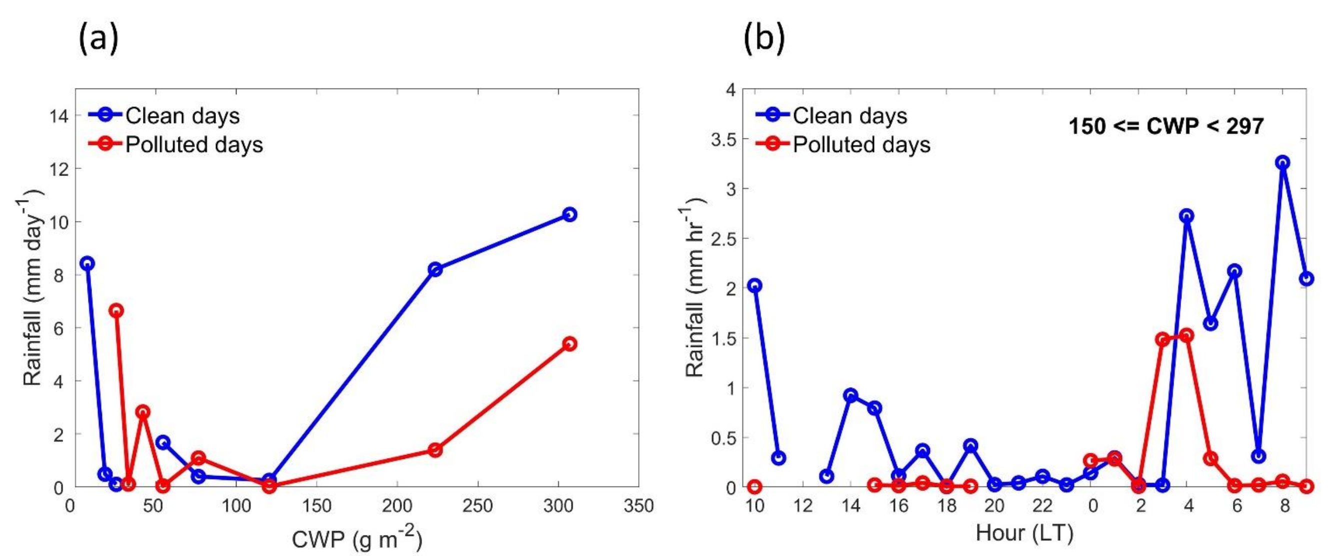



**Figure 12: Multiyear (2005-2017) (a) mean rainfall in different CWP groups calculated for clean and polluted days, and (b) ~~24 hours trend of~~ average hourly-rainfall rate calculated for clean and polluted days according to~~and considered the~~ CWP group 9 (150 ≤ CWP < 297) only. Rainfall analyses were performed from 10:00~~ am~~ and the $PM_{2.5}$ data were averaged from 10:00~~ am~~ to 14:00~~2 pm~~ as daily $PM_{2.5}$.**

**Table 1: MODIS aerosol and cloud products used in this study.**

| Product | Dataset | Acronym | Unit | Resolution |
|---|---|---|---|---|
| Aerosol (MYD04_L2, Collection 6) | Optical_Depth_Land_And_Ocean | AOD | | 10 km |
| Cloud (MYD06_L2, Collection 6) | Cloud_Effective_Radius | CER | μm | 1 km |
| | Cloud_Optical_Thickness | COT | | 1 km |
| | Cloud_Water_Path | CWP | g $m^{-2}$ | 1 km |
| | Cloud_Fraction | CF | | 5 km |
| | Cloud_Top_Pressure | CTP | hPa | 5 km |
| | Cloud_Top_Temperature | CTT | K | 5 km |
| | Cloud_Phase_Infrared | CPI | | 5 km |

**Table 2: ACI values from the literature in comparison to this study.**

| Study | ACI values | Sources | Region |
|---|---|---|---|
| Feingold et al., 2003 | 0.02-0.16 | ground-based remote sensors | Oklahoma, United States |
| Kim et al., 2008 | 0.04-0.17 | ground-based remote sensors | Oklahoma, United States |
| McComiskey et al., 2009 | 0.04-0.15 | ground-based remote sensors | California, United States |
| This study | 0.07 in CWP group 9 ($150 \leq CWP < 297$) | satellite and surface observations | Northern Taiwan |