Reply to comments on "Satellite observed indications of aerosol effects on warm cloud properties over Yangtze River Delta of China"

March 28, 2017

We would like to appreciate the reviewer for the detailed and valuable comments which helped us a lot to improve the manuscript. Our replies to all the comments are shown below.

General comments

1. Comments: (1) Why only years between 2007-2010 are considered? Given the low availability of satellite observations during these years, as it appears for example in Figures 2-3, to raw more robust conclusions would require a larger sample size. An idea could be to analyze data for the whole acquisition period of CALIPSO (i.e. since 2006).

Answer: This is correct, since the data covering the study area are only available from January 2007 to December 2010. We reanalyze all the data for the whole acquisition period between 2007 and 2010, rather than just summertime data. This issue is shown throughout the revised manuscript (all the figures were changed/modified in this respect).

2. Comments: (2) It would be beneficial to have a figure/table showing satellite retrieval availability over the analyzed domain and in all figures the sample size should be also reported.

Answer: Yes, we totally agree. We added the spatial and time series analysis of aerosol and cloud parameters over the analyzed domain in the revised manuscript, as shown in Figure 1-2 and Table 1 (see pg.9-10 in the revised manuscript). Also, we reported the sample size in all figures in the revised manuscript.

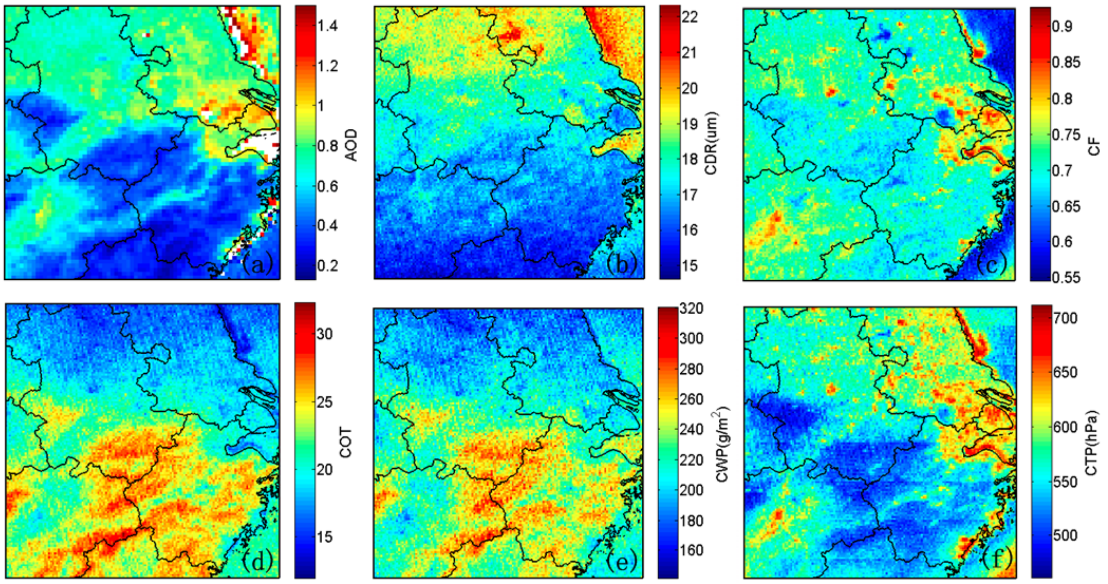

Figure 1. Spatial distributions of AOD (a), CDR (b), CF (c), COT (d), CWP (e) and CTP (f) averaged over all years between 2007 and 2010.

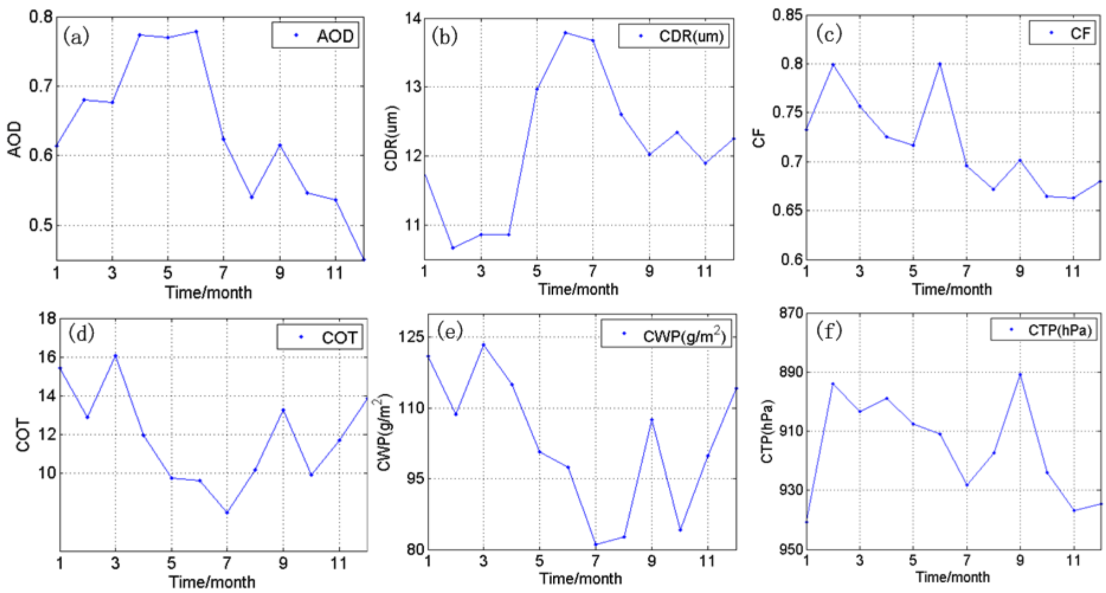

Figure 2. Time series of the monthly averaged values of AOD (a), CDR (b), CF (c), COT (d), CWP (e) and CTP (f) for all months between 2007 and 2010. Month 1 is January.

Table 1. The sample sizes of all months for each parameter

Parameters	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Total
AOD	5428	3332	3892	4704	5598	3638	5944	6630	4306	6728	6110	6400	62710
CDR	794	669	365	679	714	872	1228	2013	1514	1281	895	582	11606
COT	886	747	392	732	748	915	1298	2072	1539	1329	967	627	12232
CWP	1226	1125	620	1310	1226	1245	1490	2187	1929	1715	1261	867	16201
CF	1398	994	537	955	993	1065	1671	2650	1996	1811	1373	1119	16562
CTP	1398	994	537	955	993	1065	1671	2650	1996	1811	1373	1119	16562

3. Comments: (3) The uncertainty in the analyzed satellite retrievals should be discussed and related to the significance of the relationships identified between AOD/CDR and other cloud properties. Further, more than half of the reported correlation coefficients are either not significant or very low. I don't see a strong evidence of most of the identified relationship between the analyzed variables, based on such a small sample size, considering the uncertainty in the used retrievals and the absence of significant regression parameters.

Answer: According to these comments, we added a subsection (section 3.4 Error sources and uncertainties) into the results and discussions section (see pg.24-25). It describes uncertainties in the satellite retrievals and the significance of the relationships identified between AOD/CDR and other cloud properties. The relationships between the analyzed variables became more robust with a larger sample size, as shown in the revised manuscript. Here, we present the section 3.4 Error sources and uncertainties below.

3.4 Error sources and uncertainties

Caution is warranted in accepting the satellite-derived correlations between aerosol and cloud properties. Uncertainties in satellite data may results from assumptions on the aerosol size distribution used in the retrieval process, imperfect cloud detection resulting in residual clouds leading to high AOD values, effects of relative humidity on aerosol parameters, dynamic effect (Yuan et al., 2008). Below we analysed several potential factors that affect the interaction between aerosol and cloud below.

Firstly, the correlation between AOD and cloud parameters may be influenced by aerosol size distributions (Small et al., 2011), but MODIS retrieval does not provide aerosol size information. So it is better to explore the seasonal differences in the observed ACI due to the various aerosol emissions in different seasons. However, the relatively low number of MODIS-CALIPSO coincidences limits the further binning of the data required to further investigate this issue. Secondly, as regards cloud contamination occurrence in the AOD dataset, this is a universal and one of the most difficult problems in aerosol retrieval. Cloud detection is usually not perfect and undetected, or residual, clouds contaminate the retrieval area which leads to AOD overestimation and in turn affects the relation between aerosol and cloud properties (e.g. Sogacheva et al., 2017). A study by Mei et al. (2016), comparing their MERIS cloud mask with two independent data sets, shows that on the order of 70-90% of the cases are correctly classified as cloud free. This result is in good agreement with that from a dedicated study on consistency between aerosol and cloud retrievals from the same instrument showing that about 20% of the pixels may be mis-classified (Klueser, 2014). In this study, the samples with AOD values greater than 1.5 were excluded as a rough attempt to exclude cloud-contaminated AOD to reduce the uncertainty in the observed ACI. Thirdly, Feingold et al. (2003) reported that water vapour swelling increases the AOD. Sheridan et al. (2001) showed an important role of hygroscopic growth in determining the AOD for sea salt aerosols. The effect of humidity on the ACI has been discussed in Section 3.3.3. Finally, Young (1993) reported that ACI is influenced by dynamics through modifying radiative and thermodynamic heating.

Jones et al. (2009) emphasized the importance of vertical mixing velocity in cloud formation and ACI as discussed in Sections 3.3.4 and 3.3.5. As reported by Yuan et al. (2010), the potential artefacts above mentioned do not seem to be the primary cause for the observed relationship between aerosol and cloud parameters. Further investigations are needed to fully analyse and explain the observed phenomena.

4. Comments: (4) The way results are presented could be improved to have a more fluent and connected discussion on aerosol effects on warm clouds properties instead of presenting a description of each figure as a separate paper section. The authors should integrate all findings in a more general framework including a wider discussion on all analyzed properties and how they relate to each other.

Answer: Yes. We integrated all our findings into a more general framework, including a wider discussion on all the analyzed properties and how they are related to each other. For example, we added the sentence "Prior to investigating the aerosol impact on warm cloud properties, a general analysis of cloud properties and the effect of aerosol loading on the relations between them are discussed below." into section 3.1.2 (see pg.11 line 10-11).

The sentence "In this section we examine the responses of various cloud properties to the increasing AOD for well-separated and well-mixed clouds, respectively." was added into section 3.2 (see pg.14 lines 9-10).

The text "Based on the above findings, we conclude that in the YRD for well-mixed clouds, the CDR shows a decrease with an increasing AOD under moderately-polluted conditions, followed by an increase under polluted and heavily-polluted conditions due to the intense water vapour competition. The cloud cover behaves qualitatively similar to CDR in response to changing values of AOD. Meanwhile, cloud optical depth becomes smaller and cloud top pressure becomes larger with the increasing AOD over the whole range of AOD values" was added into section 3.2 (see pg.16 lines 20-25).

The text "Feingold et al. (2001) reported that the aerosol indirect effect depends highly on the aerosol hygroscopicity and pressure vertical velocity. Wang et al. (2014) demonstrated that the observed interaction between aerosol and cloud can be affected by the dynamical and thermodynamical processes in cloud systems. Therefore, to explore the meteorological impact on the interaction between aerosol and cloud observed over the YRD, we classify the data for various meteorological parameters, including RH (this section), LTS and PVV (Section 3.3.4)." was added into section 3.3.3 (see pg.20 lines 7-12).

We also reorganized the sentences in the conclusion section (see pg.25-27).

Specific comments

1. Comments: (1) Page 4, line 4-11: what are the spatio-temporal scales of variability of aerosol and cloud properties and how are they represented by the satellite observations you are analyzing?

Answer: The spatial and temporal variability in aerosol and cloud properties are shown in Figures 2 and 3 in the revised manuscript, respectively. We can see a decreasing north-south pattern in AOD in Figure 2a, with the highest values found in the northeast area. CDR behaves similar to AOD, except that the highest values are found in the northernmost area. Contrary to AOD, both COT and CWP show an increasing north-south pattern. Furthermore, the spatial distributions of COT and CWP are remarkably similar to each other.

The monthly-averaged values of AOD and CDR were highest in June, while December showed the lowest monthly-average value for AOD. Overall, the temporal variabilities of COT and CWP were similar, with lowest monthly averages in summer and highest averages in winter. The temporal patterns of CF and CTP were similar throughout the year (see pg.9-10).

2. Comments: (2) Page 4, line 6-7: how did you analyze "the response to the increase in aerosol loading"? Did you look at AOD temporal trends? Or do you only mean you aim at analyzing the sensitivity of cloud properties to different aerosol loading? By extending your analysis to multiple years you could also look at trends in aerosol loading (if present and if enough data are available).

Answer: We analyzed "the response to the increase in aerosol loading" in two different ways. The first one was to look at AOD temporal trends (as shown in figure 3 in the revised manuscript) and the second one was to analyze the sensitivity of cloud properties to different aerosol loading (this issue is shown through the whole manuscript). We added a spatial and time series analysis of aerosol and cloud parameters over the analyzed domain, as shown in Figure 2 and Figure 3 in the revised manuscript (see pg.9-10).

3. Comments: (3) Page 5, line 16: Why are you using Collection 5.1 instead of 6?

Answer: The MODIS Collection 05 Level 2 daily product provides cloud and aerosol properties at 10 km×10 km and 1 km×1 km (5 km×5 km) spatial resolution, respectively. The reason why we chose the MODIS Collection 5.1 is the following: most of previous researches were based on MODIS Collection 5.1 and it is easy to compare the results in our study with others' using the same data. Also, Collection 6 (C6) Aqua L2 production began in Dec 2013.

4. Comments: (4) Page 6, line 20: What is the vertical resolution of CALIOP/CALIPSO aerosol products?

Answer: The vertical resolution of the CALIOP layer product varies with altitudes: 30 m for h =0 - 8.2 km, 60 m for h = 8.2 -20.2 km, and 180 m for h = 20.2 - 30.1 km, whereas the horizontal resolution is 5 km (Liu et al., 2009). This was added into the revised manuscript (see pg.6 lines 24-26).

5. Comments: (5) Page 9, line 5: Why such few data are available in Figure 2 compared to the other figures (i.e. from Figure 4 on)? A correlation coefficient R of 0.08-0.23correspond to a coefficient of determination R^2 of 0.6-5% which indicate that your regression model is able to explain between 0.6 and 5% of the variability

in the data. Further these correlations are not significant. These results need to be better interpreted in the manuscript and the robustness of your finding to be discussed. For example it is very hard to justify that "the correlation between these parameters is negative but weak" at line 14, based on the results presented in Figure 2a. Analysis of longer time series of satellite observations may help in strengthening your conclusions. In all figures the sample size should be also reported.

Answer: All CDR, COT, CWP, CF and CTP data shown in the figures in the manuscript are averaged over AOD bins, from 0.05 to 1.5 by a step of 0.02 on a log-log scale.

However, not every CALIPSO shot has all the corresponding value for AOD, CDR, COT, CWP, CF or CTP. For example, some shots have AOD and CDR values but not COT values, while some shots have AOD and COT values but not CDR values. This reduces the sample size to some extent when considering the relationship between CDR and COT. We have reported the sample size in all the figures in the revised manuscript.

We have interpreted the results in a better way in the revised manuscript (see pg.12), and the robustness of the findings is now discussed in Section 3.4 (see pg.24-25).

6. Comments: (6) In all figures: how are the data aggregated in time? Does each dot represent a daily observation?

Answer: The time-coincidence of retrievals was assured using datasets from the same date by the A-train coordinated orbits of Aqua and CALIPSO. The detailed data preprocessing is as follows:

1. In this study, all the tracks (509) of CALIPSO covering the target study areas between 2007 and 2010 were selected. According to the latitude-longitude pairs of the profiles, all the profiles (60311) covering the target area (27°N-34°N and 115°E-122°E)were further extracted.
2. When CALIPSO shot detected the presence of aerosol, we averaged the MODIS aerosol retrievals within a radius of 50 km from the CALIPSO target. Likewise, we averaged the MODIS cloud retrievals within a radius of 5 km from the CALIPSO target. For meteorological properties, we chose the value of the footprint that was nearest to the CALIPSO target. Then, every CALIPSO shot had its corresponding AOD, CDR, COT, CWP, CF and CTP (if the value was available).
3. All CDR, COT, CWP, CF and CTP data shown in the figures in the manuscript were averaged over AOD bins, from 0.05 to 1.5 by a step of 0.02 on a log-log scale.

Therefore, each dot does not represent a daily observation in the figures in the manuscript.

7. Comments: (7) Page 9, line 8: you should include a reference describing the pollution classification based on AOD values.

Answer: We first explored the response of CDR to the increasing AOD in mixed aerosol-cloud layers and found that CDR decreases with increasing AOD in moderately polluted conditions (AOD < 0.35). In polluted and heavily polluted conditions (AOD > 0.35), however, CDR increases with increasing AOD. Here we discriminate between moderately (AOD < 0.35), polluted (AOD >= 0.35 and AOD <=0.8) and heavily polluted (AOD >0.8) conditions. These limits are somewhat arbitrary, however the AOD of 0.35 is based on analysis presented in section 3.2 where conditions change at about this value (see pg. 12).

8. Comments: (8) Page 9, line 11: this sentence needs to be rephrased. It is not clear what it means the "significance of the difference" and what the p-value refers to.

Answer: Student's t-test was used to determine whether two sets of data were significantlydifferent from each other. The p-value is defined as the probability of obtaining a result equal to or "more extreme" than what was actually observed, when the null hypothesisis true. We rephrased the sentence in the revised manuscript (see pg.12).

9. Comments: (9) Page 10: Are your results consistent with the literature? What type of significant relationship was found between COT/CWP and CDR in other studies? Given the lack of strong evidence in your results a wider discussion on what has been found so far in the literature is necessary.

Answer: In this study, we explored the response of CDR and CWP to the increasing value of COT under different pollution conditions. Costantino and Bréon (2013) compared the CDR-COT relationship of mixed and unmixed aerosol-cloud layers and found an increase in CDR with an increasing COT, followed by a decrease with higher COT in both cases (mixed and separated aerosol-cloud layers) (see pg.12 lines 24-27). They also reported that cloud water amount increases with an increasing cloud optical thickness. We added this reference into the revised manuscript (see pg.13 lines 20-21).

10. Comments: (10)Page 11, line 8: why in all panels of Figure 4 there are many more points than in Figure 2 even considering only the mixed aerosol-cloud layers?

Answer: Please see our answer to comment 5.

11. Comments: (11) Page 12, line 4: Please be more clear in explaining how you infer that "CDR is ~ 3times stronger…"

Answer: "…the relation between AOD and CDR is ~3 times stronger for the mixed layers than for separated layers…" can be inferred from the different slopes of the lines for mixed (-0.38) and separated (-0.14) layers in the previous manuscript. Now as the analyzed dataset was different, also the result changed. We rephrased the sentence in the revised manuscript (see pg.14-15).

12. Comments: (12) Page 12, line 5: the discussion of pollution levels as a function of AOD should be introduced earlier in the paper given it is used since the first analyses presented. You should also discuss why you are choosing a threshold of 0.3 instead of 0.4 and in the cited reference.

Answer: Yes, we totally agree. We modified the revised manuscript (see pg.12 lines 8-13). Now we reanalyzed all the data for the whole acquisition period between 2007 and 2010, rather than just summertime data, and we found that a threshold of 0.35 is better for differentiating between the different pollution levels over the YRD. CDR shows a negative relation with AOD in moderate polluted conditions (AOD < 0.35). In polluted and heavily polluted conditions (AOD > 0.35), however, CDR increases with an increasing AOD (see pg.12).

13. Comments: (13) Page 14, Figure 5: Why do you not separate cases with AOD> (<) 0.3 in all panels? At least in panel b there could be a different relationship if this threshold is applied.

Answer: Yes, according to these comments, we separated a case with AOD> (<) 0.35 in panel b and found a different result (see pg.14-15). However, we did not separate cases with AOD> (<) 0.35 in all the panels, as there was no significant difference in the panels c and d. Note that the threshold of AOD is 0.35 in the revised manuscript.

Technical comments

1. Comments: (1) Page 4, line 21: Figure 1 is not referenced in the manuscript, so it could be added where you introduce the analyzed domain.

Answer: Yes, we added "figure 1" where we introduce the analyzed domain in the revised manuscript (see pg.4 line 26).

2. Comments: (2) Page 6, line 7: the CALIPSO acronym needs to be defined

Answer: "…CALIPSO and CloudSat are flying in the so-called…" has been changed to "CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) and CloudSat are flying in the so-called…" in the revised manuscript (see pg.6 line 11).

3. Comments: (3) Page 8, line 2: -2 needs to be superscript

Answer: We made this change in the revised manuscript (see pg.8 line 7).

4. Comments: (4) Page 5, line 12: Since you are using only data from MODIS Aqua, the reference to the Terra satellite should be removed everywhere in the paper.

Answer: We made this change. The reference to the Terra satellite has been removed in the revised manuscript. "The MODIS sensors, onboard the Terra and Aqua satellites…" has changed to "The MODIS sensor, onboard the Aqua satellites…" (see pg.5 line 17), and "Along with the Aqua and Terra satellites…" has been changed to "Along with the Aqua satellites…" (see pg.6 line 11).

5. Comments: (5) Page 11, line 12: a space is missing between "and" and "σ"

Answer: We made this change (see pg.14 line 7).

6. Comments: (6) Page 11, line 15: remove "a" before "cloud parameters"

Answer: We made this change. "The strength of the interaction between cloud properties and AOD is quantified here as the slope of the line describing the relation between a cloud parameters and AOD, on a log-log scale, as obtained by linear regression." has changed to "The strength of the interaction between cloud properties and AOD is quantified here as the slope of the line describing the relation between cloud parameters and AOD, on a log-log scale, as obtained by linear regression." (see pg.14 line 13).

7. Comments: (7) Is there a way to differentiate the figures? Using only red and blue in all figures/panels is misleading since the reader may associate a specific color to a specific property.

Answer: Yes, we totally agree. We differentiated the figures using different markers and colors together for different cloud properties. We made this change throughout the manuscript. Here, we just take an example as shown in Fig. 3 below.

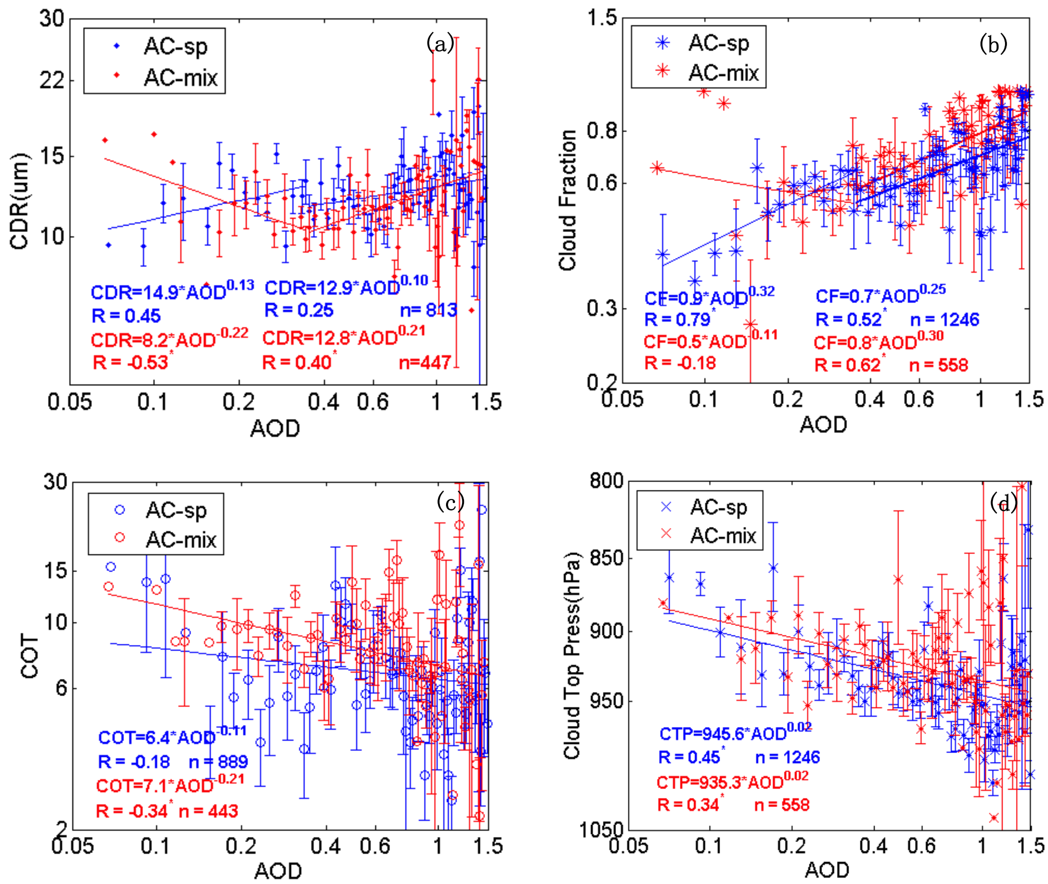

Figure 3. Scatterplots of cloud parameters versus AOD over YRD on log-log scale for cases of separated (blue) and mixed (red) aerosol-cloud layers, (a) CDR versus AOD, (b) CF versus AOD, (c) COT versus AOD and (d) CTP versus AOD. The lines present the least squares fits and the resulting relations are presented in each figure. Error bars represent the confidence level of the mean cloud parameters' value for each AOD bin, i.e. the statistical uncertainties, expressed as σ/(n-2) , where n is the number of cases within the AOD bin and σ is the standard deviation of cloud properties.

References

Feingold, G., Remer, L.A., Ramaprasad, J., Kaufman, Y.J., 2001. Analysis of smoke impact on clouds in Brazilian biomass burning regions: an extension of Twomey's approach. J. Geophys. Res. 106 (No.D19), 22907-22922.

Costantino, L. and Bréon, F.M.: Aerosol indirect effect on warm clouds over South-East Atlantic, from co-located MODIS and CALIPSO observations, Atmos. Chem. Phys., 13, 69-88, 2013.

Liu, Z., Vaughan, M., Winker, D., Kittaka, C., Getzewich, B., Kuehn, R., Omar, A., Powell, K., Trepte, C., Hostetler, C., 2009. The CALIPSO lidar cloud and aerosol discrimination: Version 2 algorithm and initial assessment of performance. J. Atmos. Ocean. Technol. 26 (7), 1198-1213.