Analysis of aerosol effects on warm clouds over the Yangtze River Delta from multi-sensor satellite observations

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Abstract. Aerosol effects on low warm clouds over the Yangtze River Delta (YRD, East China) are examined using co-located MODIS, CALIOP and CloudSat observations. By taking the vertical locations of aerosol and cloud layers into account, we use simultaneously observed aerosol and cloud data to investigate relationships between cloud properties and the amount of aerosol particles (using aerosol optical depth, AOD, as a proxy). Also, we investigate the impact of aerosol types on the variation of cloud properties with AOD. Finally, we explore how meteorological conditions affect these relationships using ERA Interim Reanalysis data. This study shows that the COT-CDR and CWP-CDR relationships are not unique, but affected by atmospheric aerosol loading. The relation between cloud properties and AOD also depends on the aerosol abundance, with a different behaviour for low and high AOD (i.e. AOD<0.35 and AOD>0.35). This applies to cloud droplet effective radius (CDR) and cloud fraction (CF), but not to cloud optical thickness (COT) and cloud top pressure (CTP). COT is found to decrease when AOD increases, which may be due to radiative effects and retrieval artefacts caused by absorbing aerosol. Conversely, CTP tends to increase with elevated AOD, indicating that the aerosol is not always prone to expand the vertical extension. Furthermore, separation of cases with either polluted dust or smoke aerosol shows that aerosol-cloud interaction (ACI) is stronger for clouds mixed with smoke aerosol than for clouds mixed with dust, which is ascribed to the higher absorption efficiency of smoke than dust. The variation of cloud properties with AOD is analysed for various relative humidity (RH) and boundary layer thermodynamic and dynamic conditions, showing that high relative humidity favours larger cloud droplet particles and increases cloud formation, irrespective of vertical or horizontal level. Stable atmospheric conditions enhance cloud cover horizontally. However, unstable atmospheric conditions favour thicker and higher clouds. Dynamically, upward motion of air parcels can also facilitate the formation of thicker and higher clouds. Overall, the present study provides an understanding of the impact of aerosols on cloud properties over the YRD. In addition to the amount of aerosol particles (or AOD), evidence is provided that aerosol types and ambient environmental conditions need to be considered to understand the observed relationships between cloud properties and AOD.

1. Introduction

Impacts of aerosols on clouds and precipitation have been reported to introduce the largest uncertainty in quantifying the anthropogenic contribution to climate change (Rosenfeld, 2000; Twomey, 1974; Gryspeerdt and Partridge, 2014; Kaufman et al., 2012). Atmospheric aerosol particles have been recognized to have two effects on Earth's climate. First, they can directly alter the energy balance due to scattering and absorption of incoming solar radiation (e.g. McCormick and Ludwig, 1967). Second, they can act as cloud condensation nuclei (CCN) and thus modify the cloud microphysical properties and lifetime as well as precipitation (Ramanathan et al., 2001; Krüger and Grassl, 2011). The effects of aerosol-induced changes of cloud properties on the radiation budget are collectively referred to as the aerosol indirect effect (AIE). The study presented here is confined to aerosol-cloud interaction (ACI) using satellite data.

The activation of aerosol particles to CCN, or more specifically the number concentration of CCN, is a direct link between aerosols and clouds, and the aerosol activation efficiency is a key aerosol property affecting aerosol-cloud interaction. For a given constant cloud liquid water path (CWP), an increased aerosol loading is expected to lead to smaller and more numerous cloud droplets, resulting in an increase of cloud albedo. This process, termed as the "first AIE" or "Twomey's effect", may lead to a net cooling of climate (Twomey, 1974; Feingold et al., 2003). The reduced cloud droplet effective radius (CDR) also suppresses precipitation and can consequently increase cloud lifetime, thus maintaining a larger liquid water path, with a possible further increase in the cloud optical thickness (COT) and cloud reflectance. This process, described as the "second AIE", may further influence the cloud fraction (CF) (Albrecht, 1989; Feingold et al., 2001). The interaction mechanisms between aerosols and clouds remain among the most uncertain processes in the global climate system in spite of a large number of studies made using both observations (Platnick et al., 2003; Koren et al., 2005) and models (Suzuki et al., 2004; Quaas et al., 2009; Sena et al., 2016).

In order to better understand aerosol indirect effects, we resorted to statistical analysis of satellite observations. By virtue of their large coverage and high spatial and temporal resolution, satellite-borne instruments have become a promising observational tool in studying aerosol-cloud interactions. Previous studies using a large amount of satellite data and/or multiple satellite instruments have shown that aerosol particles can affect cloud properties significantly (Krüger and Grassl, 2002; Menon et al., 2008; Sporre et al., 2014; Rosenfeld et al., 2014; Saponaro et al., 2017). Satellite measurements suggest that the CDR tends to decrease with increasing aerosol loading, which is consistent with Twomey's theory (Matheson et al., 2005; Meskhidze and Nenes, 2010; Koren et al., 2005). However, also positive correlations between CDR and aerosol optical depth (AOD) have been found in some study areas, from both observations and models, especially over land (Feingold et al., 2001; Grandey and Stier, 2010; Yuan et al., 2008). Different behaviours of CDR as function of AOD for different AOD regimes (low/high) have been observed by, e.g., Tang et al. (2014) and Wang et al. (2015). Feingold et al.(2001) concluded that there are three kinds of CDR responses to aerosol enhancement: the CDR decreases with increasing aerosol loading followed by (1) a saturation of the value of CDR in response to high AOD, (2) a decrease in the CDR with further increasing AOD due to suppression of cloud water vapour supersaturation caused by abundant large particles, or (3) an increase in CDR with further increases in AOD due to an intense competition for vapour which evaporates the smallest droplets. Likewise, the aerosol impact on COT is still poorly quantified. Costantino and Bréon (2013) reported that the relationship between AOD and COT, which can be either positive or negative, depends on the balance between the simultaneous CDR increase and CWP decrease when AOD increases. With regard to the impact of aerosols on the cloud life cycle, it is of great importance to explore the relationship between the aerosol loading and cloud fraction, because the cloud fraction is highly associated with other cloud properties and has a large effect on radiation (Gryspeerdt et al., 2016). Kaufman (2006) and Koren (2008) reported an increase in the cloud cover with an increasing aerosol loading, followed by an inverse pattern due to the absorption efficiency of aerosol. This brief summary shows that the aerosol effect on cloud properties and the magnitude of this effect are still very unclear.

Aerosol and cloud properties may have different vertical distributions and may actually not physically interact. Costantino and Breon (2013) and Jones et al. (2009), using MODIS data, found that the aerosol indirect effect is stronger for well-mixed clouds than for well-separated clouds (in well-mixed aerosol and cloud layers are physically interacting, as further explained in Section 2).These observations show that it is important to consider the relative altitudes of aerosol and cloud layers when estimating the aerosol indirect effects. In addition, local differences in aerosol populations and cloud regimes may have a strong effect on ACI (Sinha et al., 2003; Small et al., 2011; Kaufman et al., 2005b). Yuan et al. (2008) proposed that the chemical composition of aerosol particles may play a role in determining the relationship between AOD and CDR. Meteorology can affect the interaction between aerosol and cloud, which usually further complicates ACI (Koren et al., 2010; Reutter et al., 2009; Loeb and Schuster, 2008; Su et al., 2010; Stathopoulos et al., 2017). As a consequence, the widely varying estimates of the aerosol impact on cloud parameters, either positive or negative, depend on factors like the aerosol size distribution and chemical composition, cloud regime and local meteorological conditions. Therefore, the dataset used in this study contains not only aerosol and cloud properties derived from MODIS, CALIOP and CloudSat, but also the meteorological parameters collected from the daily ERA Interim Reanalysis data.

The Yangtze River Delta (YRD) is characterized by a variable aerosol composition and increasing aerosol concentration during the last two decades (Ding et al., 2013a; Qi et al., 2015). Using multi-sensor retrievals, this study aims to systematically examine the response of warm cloud parameters (CDR, CF, COT and CTP) to the increase in the aerosol loading, where AOD is used as a proxy for CCN number concentration (Andreae, 2009; Kourtidis, et al., 2015). New insights into the changing cloud properties over a wide range of aerosol loadings, in particular in high AOD conditions, result from our focus on a systematic understanding of ACI from three perspectives: (1) well-mixed and well-separated clouds, (2) aerosol effects on properties of well-mixed clouds, (3) well-mixed clouds under different meteorological conditions.

The paper is organized as follows: section 2 describes the datasets used, data processing and the main analysis conducted to explore aerosol cloud interaction. Section 3 starts with a general description of aerosol and cloud properties and the effect of aerosol loading on the relations between them, followed by a description of aerosol effects on cloud properties (CDR, CF, COT and CTP). In the latter we discriminate between well-separated and well-mixed clouds. The focus will be on well-mixed clouds where ACI takes place, and aerosol types and meteorological factors are considered to better understand the possible mechanisms. Overall conclusions and discussions are presented in section 4.

2. Methods

2.1 Description of the study region

In this study, the Yangtze River Delta (YRD), covering the area 27°N-34°N and 115°E-122°E (Figure 1), was chosen in order to investigate the aerosol-induced variability in micro- and macrophysical properties of low warm clouds during four consecutive years (2007-2010). The YRD region was chosen because it is representative for the continental East Asian subtropical climate. The marine monsoon subtropical climate for YRD is characterized by hot and humid summers and cool dry winters (Sundström et al., 2008; Zhang et al., 2010). The mean temperature in summer from 2007 to 2010 is about 27-28℃. Mean annual precipitation ranges from 1000 to 1400 mm and most precipitation occurs in spring and summer (Zhang et al., 2010; Cao et al., 2016).

The population density in the YRD is very high with intensive human activities in the region contributing to a very variable and complex aerosol composition. The YRD has been reported to be a major source region of both black carbon and sulfate (Wang et al., 2014; Andersson et al., 2015). In addition, other aerosol sources such as dust emissions render the interactions between aerosols and clouds complicated (Nie et al., 2014). The continental area of interest is characterized by a high level of anthropogenic emissions and is well suited for research related to the indirect effects of aerosols on cloud micro- and macro-physical properties.

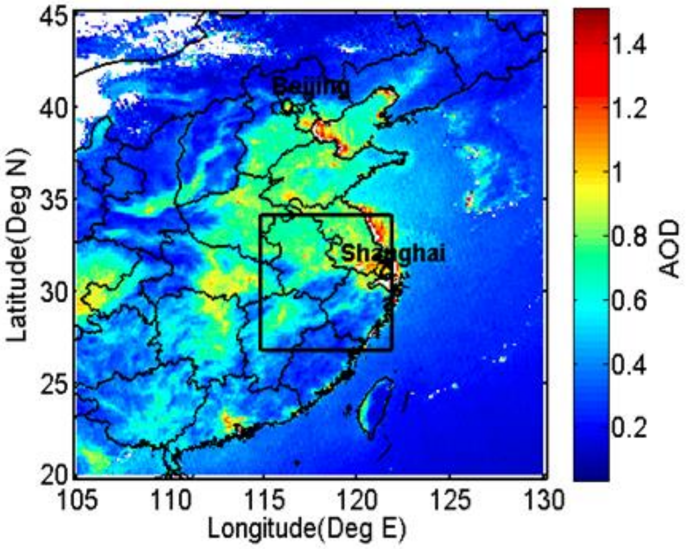

Figure 1. Map of annual averaged MODIS/AQUA level 2 AOD for all years during the period from 2007 to 2010. The black rectangle (27°N-34°N and 115°E-122°E) indicates the Yangtze River Delta (YRD).

2.2 Data sources

The MODIS sensor, onboard the Aqua satellite, has a swath width of ~2300 km and multi-band spectral coverage (King et al., 2003). The MODIS/Aqua overpass time for the study area is around 13:30 local time, when continental warm clouds are likely to be well developed. Therefore MODIS/Aqua was selected as a data source to explore the ACI over this area. In this work, we used the MODIS Collection

5.1 AOD product (MOD04) derived from cloud-free pixels (resolution 500 m at nadir) and aggregated to a resolution of 10 km×10 km (Remer et al., 2005; Levy et al., 2010). The AOD over land is retrieved using three MODIS channels: 0.47, 0.66 and 2.13 µm (Remer et al., 2005). Cloud properties are retrieved using six spectral channels (King et al., 1998) at visible and near infrared wavelengths (i.e., 0.66, 0.86, 1.24, 1.64, 2.12 and 3.75 µm). Here, we used the AOD as a proxy for aerosol burden in our aerosol-cloud interaction analysis. The cloud properties used in this study, CDR, CWP, COT, cloud top pressure (CTP) and cloud phase infrared (CPI), were obtained from the Level 2 cloud product (MYD06) (King et al., 2003). Both these products MOD04 and MYD06 are in good agreement with ground-based remote sensingdata (Levy et al., 2010; Platnick et al., 2003). More detailed information on algorithms for the retrieval of aerosol and cloud properties is provided at http://modis-atmos.gsfc.nasa.gov.

Along with the Aqua satellites, CloudSat and CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) are flying in the so-called "A-train" constellation together with other NASA satellites (Stephens et al., 2002). CloudSat carries the CPR (Cloud Profiling Radar), i.e. the first satellite-based millimeter-wavelength cloud radar to detect the vertical information on different sized cloud droplets (Im et al., 2005). The CPR is able to penetrate optically thick clouds and detect weak precipitating particles (Wang et al., 2013). In the present study we utilized the datasets CloudLayerBase and CloudLayerTop from 2B-CLDCLASS-LIDAR, the latest version (R04) of the CloudSat standard data products. The data are provided in the CPR spatial grid with vertical and horizontal resolutions of approximately 480 m and 1.4×1.8 km, respectively. CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) on board CALIPSO is the first space-borne near-nadir polarization lidaroptimised for aerosol and cloud measurements (Winker et al., 2003). It is sensitive to optically thin clouds which could be missed by CPR (Wang et al., 2013). The datasets Layer_Base_Altitude and Layer_Top_Altitude retrieved from the CALIOP level-2 aerosol layers product (05kmALay) were used in the present study. Its footprint is very narrow, with a laser pulse diameter of 70 m on the ground. The vertical resolution of the CALIOP layer product varies with altitude: 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). Combining CloudSat and CALIPSO observations has provided new insights into the vertical structure and microphysical properties of clouds (Matrosov, 2007).

The daily temperature at the 1000 hPa and 700 hPa levels, relative humidity at the 950hPa level and pressure vertical velocity at the 750 hPa level were obtained from ERA Interim Reanalysis data. The daily ERA Interim Reanalysis contains global meteorological conditions with 0.125°×0.125° grids and a 37 level vertical resolution (1000-1 hPa) every six hours (00:00, 06:00, 12:00, 18:00 UTC) (http://apps.ecmwf.int/datasets/data/interim-full-daily/). The reanalysis data were used for the closest collocation with the satellite overpass time over the study area.

Table 1. Level 2 MODIS, CALIOP, CALIOP/CPR and ERA Interim products used to characterize aerosol and cloud properties.

Product	**Dataset**	**Horizontal resolution**	**Data source**
Aerosol(MYD04 Level 2 Collection 5)	Optical_Depth_Land_And_Ocean	10 km	MODIS
Cloud(MYD06 Level 2 Collection 5)	Cloud_Effective_Radius	1 km	
	Cloud_Water_Path	1 km	
	Cloud_Phase_Infrared_Day	5 km	
	Cloud_TOP_Pressure_Day	5 km	
	Cloud_Fraction_Day	5 km	
	Cloud_Optical_Thickness	1 km	
Cloud(2B-CLDCLASS-LIDAR)	CloudLayerBase	2.5 km	CALIOP/CPR
	CloudLayerTop	2.5 km	
Aerosol(05kmALay)	Layer_Top_Altitude	5 km	CALIOP
	Layer_Base_Altitude	5 km	
	Cloud_Aerosol_Discrimination	5 km	
	Feature_Classification_Flags	5 km	
ERA Interim	Temperature (700hPa, 1000hPa)	0.125°	ECMWF
	Relative humidity (950hPa)	0.125°	
	Pressure vertical velocity (750hPa)	0.125°	

2.3 Data processing

The MODIS/AQUA, CALIOP/CALIPSO and CPR/CLOUDSAT satellites are part of the A-Train constellation and observe the same scene on Earth within one to two minutes (Stephens et al., 2002). Therefore, time-coincidence of retrievals is assured when the datasets are extracted for the same date. Meteorological properties retrieved from the 06:00 UTC ERA Interim datasets were used here as the “A-train” satellites constellation overpasses the region of interest at about 13:30 local time (05:30 UTC). We aggregated CDR, COT and CWP (1 km × 1 km) to a resolution of 5 km × 5km to match the along-track resolution of CALIOP (5 km × 5 km), while CTP, CF and CPI were directly applied for the analysis since all of them are at a 5 km × 5km spatial resolution.

Aerosol properties are only retrieved for strictly cloud-free pixels as determined by the application of a cloud-detection scheme. However, cloud detection schemes are not perfect and some residual clouds may remain undetected resulting in high AOD (Kaufman et al., 2005b). Another potential source of error could be the misclassification of high AOD areas, such as in the presence of desert dust or very high pollution levels, as clouds. To reduce a possible over-estimation of AOD, cases with AOD greater than 1.5 were excluded from further analysis. In this paper, we focused on warm clouds with CTP larger than 700 hPa and CWP lower than 200 g m^{-2}, as most aerosols exist in the lower troposphere (Michibata et al. 2014). In addition, only cases with CPI = 1 (liquid water cloud) were included. When CALIOP detected the presence of aerosol, we averaged the MODIS aerosol retrievals within a radius of 50 km from the CALIOP target. Likewise, we averaged the MODIS cloud retrievals within a radius of 5 km from the CALIOP target. For meteorological properties, we chose the value of the footprint that is nearest to the CALIOP target. MODIS, CALIOP, and CPR datasets are listed in Table 1.

A quantitative relationship between aerosol optical depth and cloud properties has been documented in previous studies (Sporre et al., 2014; Meskhidze and Nenes, 2010; Koren et al., 2005, Saponaro et al., 2017). However, the relative vertical positions of aerosol and cloud layers contribute to the uncertainty in this relationship. Following the method by Costantino and Breon (2013), we considered the aerosol and cloud layers to be physically interacting (well mixed) when the vertical distance between bottom (top) of the aerosol layer and the top (bottom) of a cloud layer was smaller than 100 m. Coincident samples with a vertical distance larger than 750 m were assumed to be “well separated”. Coincident samples with a distance between 100 and 750 m were defined as “uncertain”. The uncertain cases, as identified using the information from CloudSat, were excluded from further analysis in this study. Cloud types were identified as single-, double- and multi-layer clouds using the cloud layer information at each point. Single-, double- and multi-layer cloud samples accounted for 59 %, 30% and 11 % of the total samples, respectively. Using the highest occurrence frequency (OF) of aerosol type below 10 km altitude at each point, the aerosol type of highest OF was defined following the Feature_Classification_Flags derived from CALIOP.

Meteorological and aerosol impacts on cloud macrophysics and microphysics are found to be tightly intermingled (Stevens and Feingold, 2009). In an attempt to isolate aerosol effects, the meteorological effects on clouds were explored in a statistical sense. Meteorological properties used here include relative humidity (RH), lower tropospheric stability (LTS) and pressure vertical velocity (PVV). LTS is defined as the difference in potential temperature between the free troposphere (700hpa) and the surface, which is representative of typical thermodynamic conditions (Klein and Hartmanm, 1993). RH, LTS and PVV have been suggested to affect aerosol and cloud interaction (Gryspeerdt and Partridge, 2014; Small et al., 2011). A positive LTS is associated with a stable atmosphere in which vertical mixing is prohibited; negative PVV indicates a local upward motion of air parcels.

3. Results and Discussions

3.1 Overall aerosol and cloud characteristics

3.1.1 Spatial and time series analysis of aerosol and cloud parameters

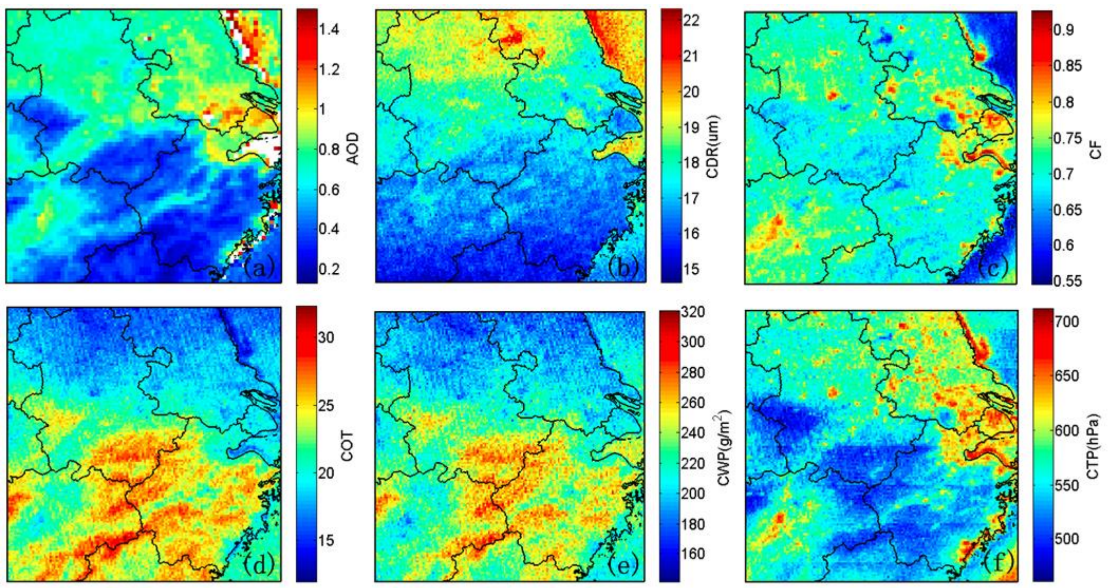

Figure 2. Spatial distributions of AOD (a), CDR (b), CF (c), COT (d), CWP (e) and CTP (f) averaged over all years between 2007 and 2010.

The spatial variations of the aerosol and cloud properties over the study area, averaged over the years 2007-2010, are shown in Figure 2. 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.

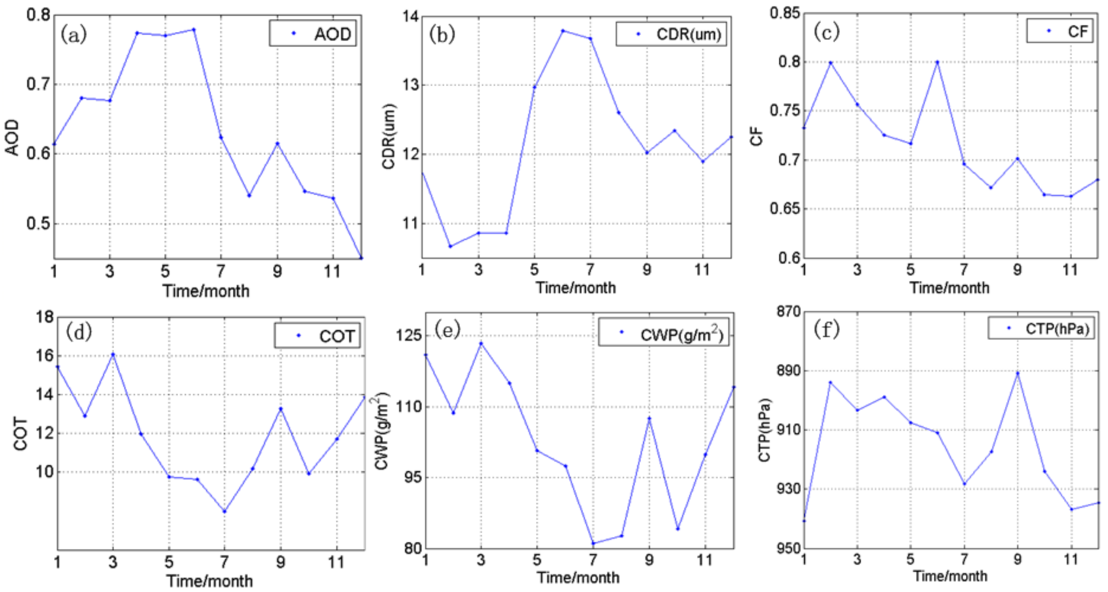

Figure 3. Time series of the monthly averaged values of AOD (a), CDR (b), CF (c), COT (d), CWP (e) and CTP (f) for all months between 2007 and 2010. Month 1 is January.

Figure 3 shows time series of the monthly-averaged values for the AOD, CDR, COT, CWP, CF and CTP, calculated for each month during the four years 2007 - 2010. Both the monthly-averaged AOD and CDR are highest in June. December presents the lowest monthly-average for the AOD. Overall, the variations of the monthly-averaged COT and CWP are similar, with the lower values in the summer and the higher value in the winter. The monthly-averaged CF approaches its maximum values in Jan and June, while CTP shows two peaks in Feb and Sep. Note that CTP is plotted along the vertical axis from high to low. The monthly averages are determined from the numbers of samples presented in Table 2 for each parameter and each month between 2007 and 2010. Further, the availabilities of data for AOD and cloud properties are not the same for the whole acquisition period between 2007 and 2010. It indicates that not every CALIPSO shot has all the corresponding value for AOD, CDR, COT, CWP, CF or CTP, which will decrease the data sample size to some extent.

Table 2. 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

3.1.2 Variation of COT and CWP with CDR

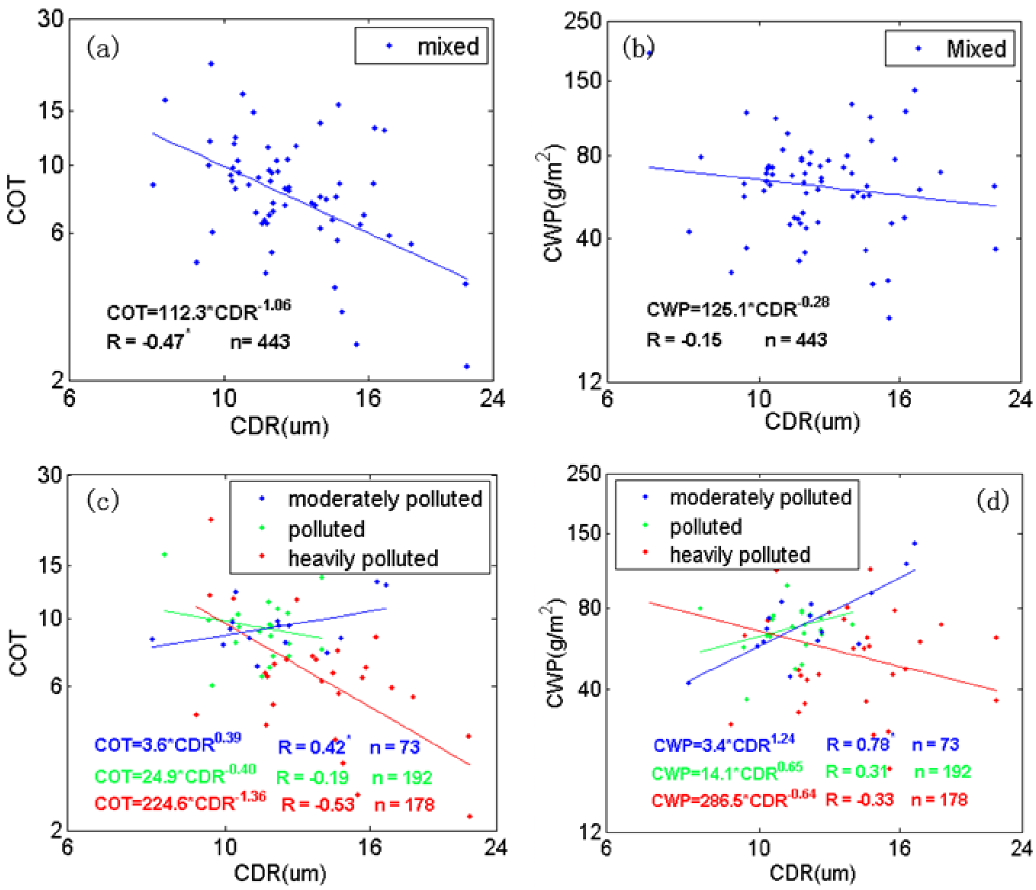

Figure 4. Scatterplots of cloud parameters versus CDR in well-mixed aerosol-cloud layers: (a) COT and (b) CWP, both for all data; (c) COT and (d) CWP, both for data grouped by moderately polluted (in blue), polluted (in green) and heavily polluted (in red) atmospheric conditions. Here moderately polluted refers to AOD <0.35, polluted refers to 0.35<=AOD <= 0.8 and heavily polluted refers to AOD>0.8. The lines present the least squares fits and the resulting relations are presented in each figure. The number of data samples is also reported in the figure (and following figures).

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. The overall statistical relations between the cloud parameters used in this study are derived from the scatterplots shown in Figure 4. All CDR, COT, CTP and CWP data shown in Figure 4 (and later figures) are averaged over AOD bins, from 0.05 to 1.5 with a step of 0.02 on a log-log scale. Student's t-test is used to determine whether two sets of data are significantly different 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 hypothesis true. Marker * at the top right corner of R-value denotes statistically significant if $p<0.05$.

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. The threshold of 0.35 for AOD is chosen based on analysis presented below in section 3.2 where we compare the relation of cloud parameters and AOD in more detail. Figure 4(a) shows a scatterplot of COT vs CDR for well-mixed clouds. The correlation between these parameters is negative, i.e. COT decreases with CDR, with a correlation coefficient equal to -0.47. Figure 4(c) shows the same data but distinction is made between data points with in moderately polluted, polluted and heavily polluted conditions. For this dataset, COT increases with an increasing CDR at moderately polluted conditions. In contrast, for heavily polluted conditions COT shows a decrease with an increasing CDR. This may indicate the existence of intense competition between the aerosol particles for water vapour where the larger droplets are more prone for condensation of water vapour than smaller ones, and thus grow to larger sizes. This results in a shift of the droplet spectrum to larger sizes due to the increase of CDR accompanied by a decrease of COT (Wang et al., 2015). The data for the three different AOD cases show that the relationship between CDR and COT is not unique and depends on the aerosol abundance. Costantino and Bréon (2013) compared the CDR-COT relationship of mixed and separated aerosol-cloud layers and found an increase in the CDR with increasing COT, followed by a decrease with higher COT in both cases (mixed and separated aerosol-cloud layers). Compared to their study, we consider the effect of aerosol loading on the relationship between CDR and COT in both cases.

Figure 4(b) shows a weak correlation between CWP and CDR for well-mixed cloud layers, with a correlation coefficient equal to -0.15. However, when different degrees of pollution are considered, Figure 4(d), we see a clear correlation between both parameters (R=0.78) in moderately polluted conditions, where CWP clearly increases with increasing CDR. In polluted and heavily polluted conditions the variation of CWP with increasing CDR is much weaker, R=0.31 for polluted conditions, and in heavily polluted conditions CWP decreases with increasing CDR (R=-0.33).

3.1.3Variation of COT and CWP with cloud top height

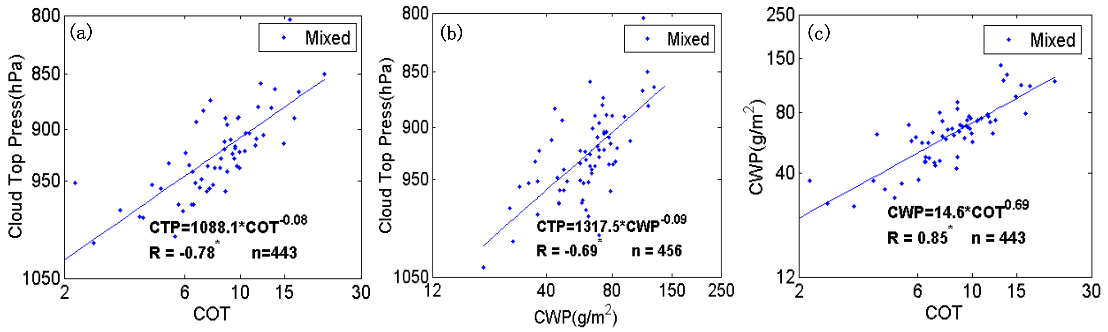

Figure 5. Scatterplots of cloud parameters in well-mixed aerosol cloud layers for all data: (a) CTP versus COT, (b) CTP versus CWP, (c) CWP and COT. The lines present the least squares fits and the resulting relations are presented in each figure.

CTP is generally used as a measure of cloud top height (CTH), with higher CTP implying a lower CTH. Figure 5(a) shows a positive correlation between CTP and COT, implying the occurrence of higher clouds with an increasing COT, which is consistent with the general understanding of aerosol-cloud interactions. Note that here and in the following figures, CTP is plotted along the vertical axis from high to low, i.e. decreasing CTP indicates increasing CTH, and positive correlations between CTP and other cloud parameters indicate that an increase in these parameters corresponds to a higher CTH. Figure 5(b) shows a positive correlation between CTP and CWP, which again implies that clouds are higher as CWP increases. An explanation for this phenomenon is provided by Gao et al. (2014), i.e. clouds grow in the vertical and more drizzle is produced, so that the cloud liquid water path becomes larger. Figure 5(c) shows the relation between CWP and COT. The CWP increases with the increase of COT, which is in good agreement with the aerosol second indirect effect hypothesis that the precipitation suppression can increase CWP and possibly further increase COT. This observation is in good agreement with those of Costantino and Bréon (2013) that cloud water amount increases with increasing cloud optical thickness.

3.2 Difference between separated and mixed conditions

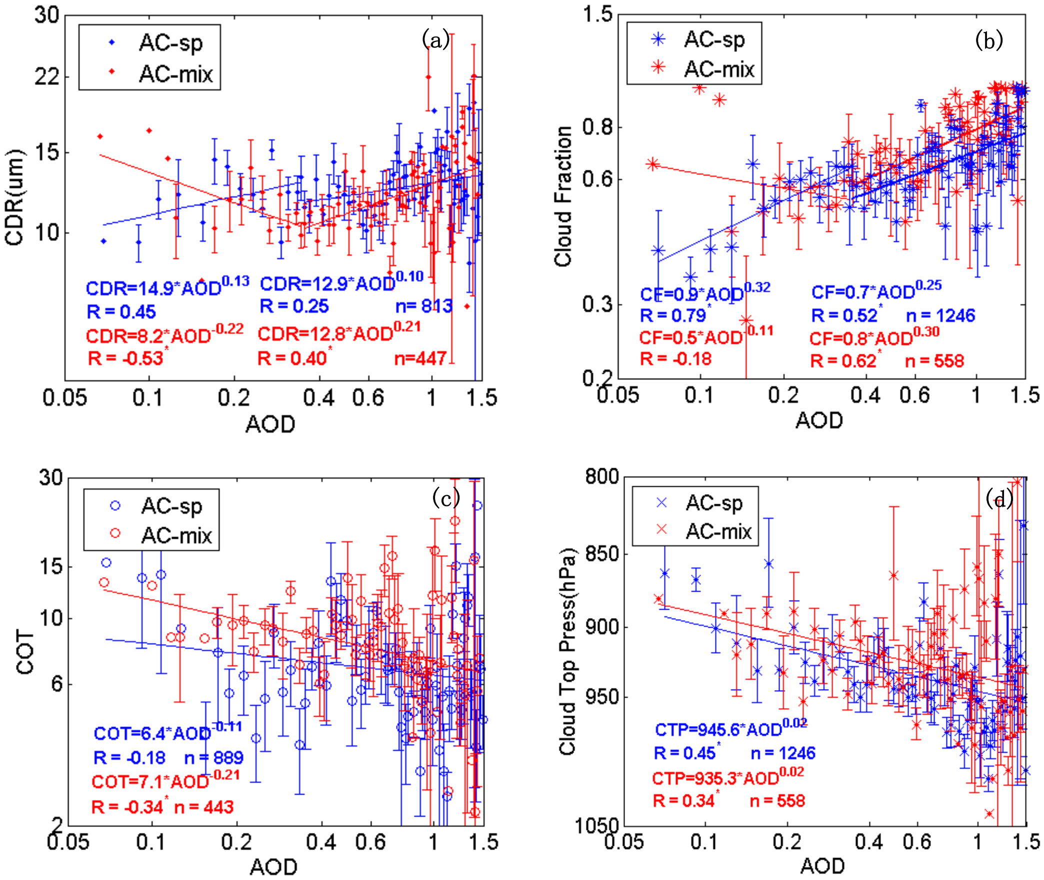

Figure 6. Scatterplots of cloud parameters versus AOD over YRD on log-log scale for cases of separated (blue) and mixed (red) aerosol-cloud layers, (a) CDR versus AOD, (b) CF versus AOD, (c) COT versus AOD and (d) CTP versus AOD. The lines present the least squares fits and the resulting relations are 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.

In this section we examine the responses of various cloud properties to the increasing AOD for well-separated and well-mixed clouds, respectively. Figure 6 shows relations between cloud parameters (CDR, CF, COT, CTP) and AOD for both separated and mixed conditions. 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. In figure 6(a), CDR shows a negative relation with AOD in moderately polluted conditions when aerosol and cloud layers are mixed, which is in good agreement with Twomey's theory (Twomey, 1977). We note that, due to the limited number of data points in the dataset with AOD < 0.35, the present work does not allow selecting conditions with a constant CWP. Following, e.g., Costantino and Breon (2010; 2013) and

Wang (2015) we use all available data together. In polluted and heavily polluted conditions, however, CDR increases with increasing AOD, suggesting some sort of saturation in aerosol-cloud interactions when AOD approaches 0.35. This value for the tipping point (0.35) is close to the value of 0.4 reported by Feingold et al. (2001). As discussed earlier, Feingold et al. (2001) proposed three primary responses of CDR to the aerosol loading. We consider the fact that CDR increases with an increase in AOD when AOD loading exceeds 0.35 as "anti-Twomey effect". The positive relation between CDR and AOD may be similar to that described by Feingold et al. (2001), case 3 (see above), i.e. due to intense vapour competition the smaller droplets evaporate as the number of particles continues to increase. It may also be that only a subset of aerosol particles is activated when not enough vapour is available, and once activated they continue to grow faster, thus preventing water vapour from condensing onto smaller aerosol particles that are less susceptible to activation, resulting in the increase of CDR.

Figure 6a also shows that in well-separated cloud layers CDR varies much less with AOD irrespective of whether the AOD is relatively low or high. Such a weaker variation can be attributed to the fact that no aerosols are subjected to cloud microphysical process since there are no physical interactions between aerosol and cloud layers.

Figure 6(b) shows that when aerosol and cloud layers physically interact, the CF shows a decrease with an increasing AOD in moderately polluted conditions, albeit with a low significance as indicated by the small correlation coefficient R, followed by an inverse patternin polluted and heavily polluted conditions. This outcome is not in agreement with the findings of Koren et al. (2008) and Small et al. (2011). It could be explained as follows: Here, when aerosol and cloud layers are well-mixed, the absorption of solar radiation heats the mixed layer and reduces the cloud cover due to the quite high concentrations of the smoke particles over the YRD. This feedback would be balanced once the heating of the surface raises the surface temperature. It destabilizes the atmosphere resulting in vertical transport and thus enabling transfer of humidity from the surface to higher levels in the atmosphere. This effect increases cloudiness (Koren et al., 2008). Conversely, CF shows an increasing pattern with an increasing AOD for the whole AOD dataset in well-separated cloud layers. This increase might be due to absorbing aerosols interacting with incoming solar radiation above the cloud layer (Costantino and Bréon, 2013). In this process, absorbing aerosols above cloud top may heat the aerosol layer and cool the surface, thereby stabilizing the boundary layer and maintaining a moist boundary layer. In addition, scattering aerosol reduces the amount of solar light reaching the surface. This combination of two effects suppresses cloud vertical development and increase the low cloud cover.

The COT has a negative correlation with AOD in both conditions, as shown in Figure 6(c). There are two effects that may contribute to this negative relationship. On one hand, the evaporation of cloud droplets caused by locally absorbing aerosol makes clouds thinner, which is a radiative effect. On the other hand, the presence of absorbing aerosol may influence the satellite-retrieved COT because it can absorb radiation and thus reduce the cloud reflectance measured by the sensors on the satellite (Meyer et al., 2013; Li et al., 2014; Meyer et al., 2015; Hoeve et al., 2011). Meyer et al. (2013) reported that adjusting for above-cloud aerosol attenuation can increase the retrieved regional mean COT by roughly 18% for polluted marine boundary layer clouds. Li et al. (2014) also found that,due to absorbing aerosols in the heart of the Yangtze Delta region, satellite observations tend to underestimate COT. The radiative effect and retrieval uncertainty could be the important factors for the decrease of COT with increasing AOD, as suggested by Hoeve et al. (2011) and Alam et al. (2014). These authors reported similar results on the decrease of COT with increasing AOD, which may result from the measured reflectance from a cloud top at wavelengths in the visible being smaller than expected due to absorbing aerosols.

The relationship between CTP and AOD has been plotted in Figure 6(d). There is a positive correlation between CTP and AOD, which is contradicting the general understanding that high aerosol loading will result in an increase of cloud lifetime and higher cloud top. The positive relation between CTP and AOD has an implication that higher aerosol abundance is not always accompanied by smaller cloud top pressure. This suggests that the primary effect of aerosol is not always to produce taller and more convective clouds in some cases (Rennóet al., 2013).

Based on the above findings, we conclude that for well-mixed clouds in the YRD, 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 increasing AOD over the whole range of AOD values.

3.3 Case of mixed aerosol-cloud layers

3.3.1 ACI for single-layer mixed clouds

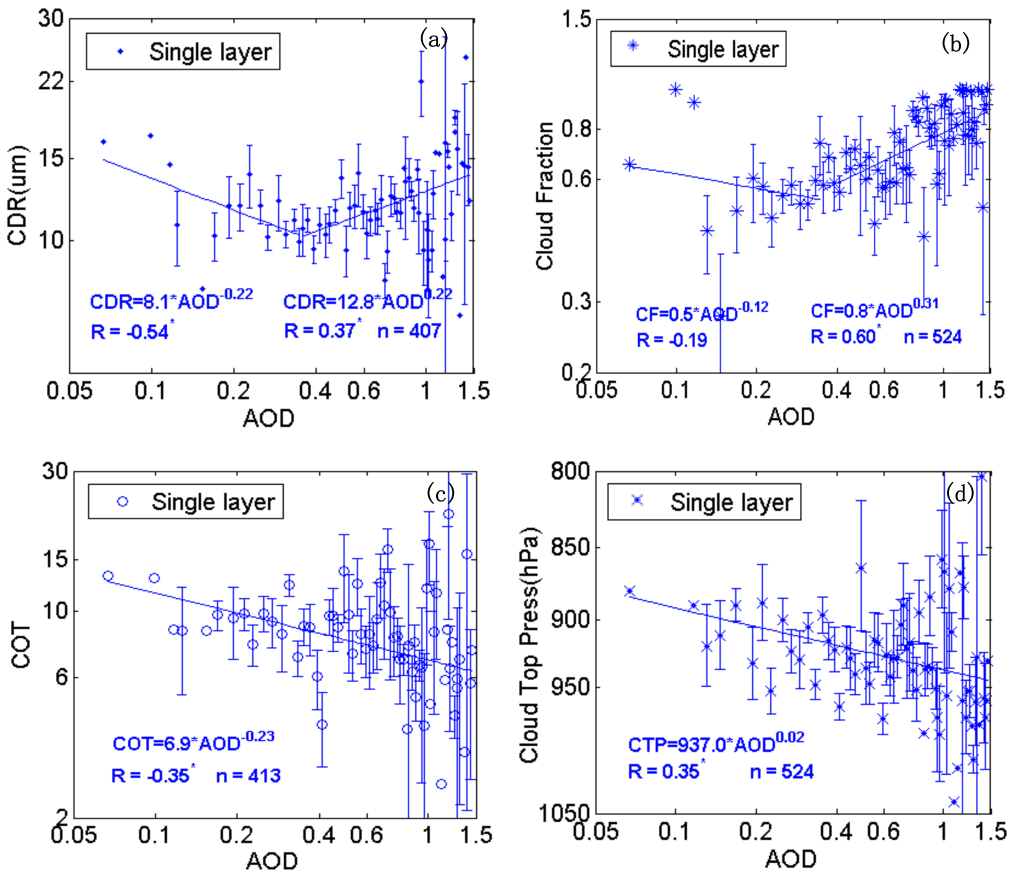

Figure 7. Scatterplots of cloud parameters versus AOD over YRD on log-log scale for mixed aerosol-single layer clouds, (a) CDR, (b) CF, (c) COT and (d) CTP. The lines present the least squares fits and the resulting relations are presented in each figure. The error bars indicate the statistical uncertainties as in Fig. 6.

Well-mixed clouds show a stronger relation between aerosol and cloud properties than separated clouds, as shown above. From here on, we will focus on potential aerosol indirect effects on well-mixed warm clouds as defined above. Relations between CDR, CF, COT and CTP with AOD will be explored in this section. Figure 7 shows the variation of single layer cloud properties with AOD when aerosol and cloud layers are mixed. The relation between CDR and AOD changes from negative for AOD < 0.35 to positive for AOD > 0.35 (Figure 7a). As with the CDR, the CF shows similar variation with the elevated AOD over the whole AOD range. Figure 7(c) shows COT is negatively associated with increasing of AOD. In contrast, CTP decreases with increasing AOD (Figure 7d), i.e. cloud top height increases. In general, the characteristics for cases of mixed aerosol-single layer warm clouds (Figure 7) are quite similar to the case of mixed aerosol-warm clouds (Figure 6). The slightly difference of fits comes from the different types of clouds are considered in different conditions. In fig 6, the clouds are not limited to single layer warm clouds, but also double layer warm clouds.

3.3.2 Influence of aerosol type on ACI

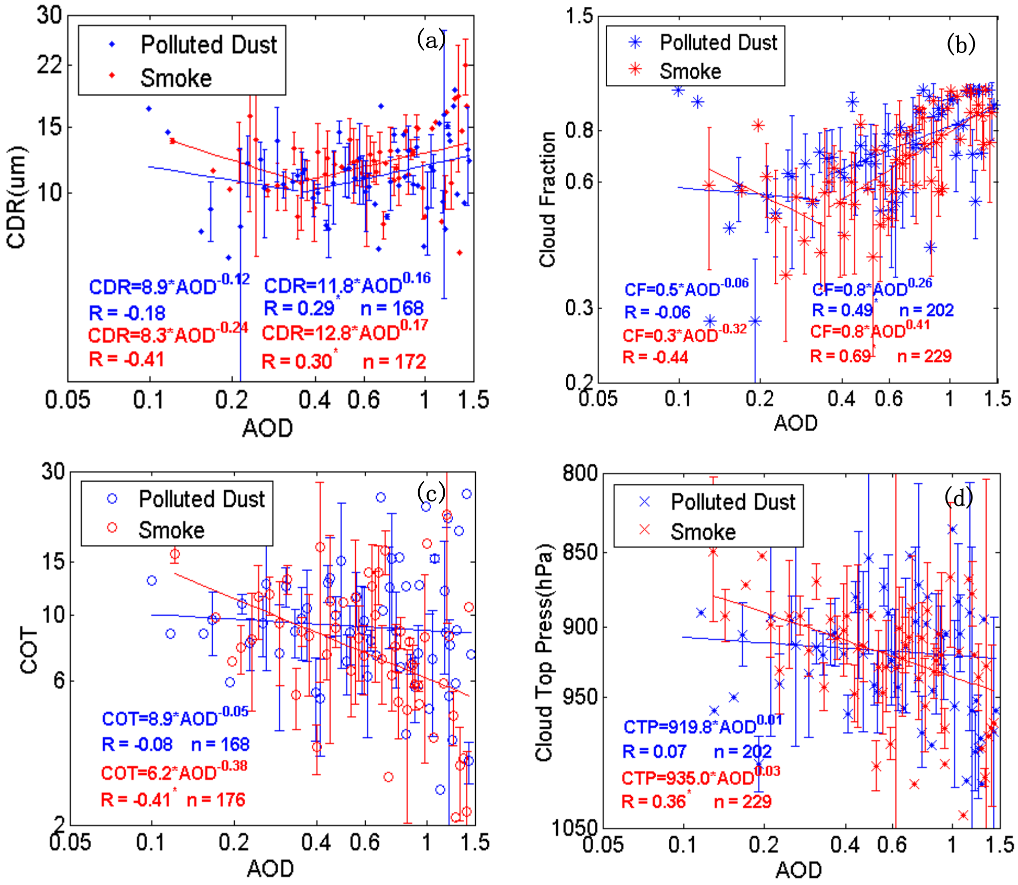

Figure 8. Scatterplots of cloud parameters versus AOD over YRD on log-log scale for cases of mixed dust aerosol-cloud layers (blue) and mixed smoke aerosols-cloud layers (red), (a) CDR, (b) CF, (c) COT and (d) CTP. The lines present the least squares fits and the resulting relations are presented in each figure. The error bars indicate the statistical uncertainties as in Fig. 6.

Eastern China is a region with high concentrations of sulfate, dust, black carbon and other carbonaceous aerosols. In heavily polluted areas, dust aerosols become coated with hygroscopic material, making them effective CCN (Levin et al., 1996; Satheesh et al., 2006). Especially, there are high emissions of smoke by strawburning in summertime. Aerosol - cloud interaction is strongly dependent on the aerosol types, their size distribution and the vertical variation of these, as well as ambient environmental conditions (Patra et al., 2005; Matsui et al., 2006; Dusek et al., 2008; Yuan et al., 2008). Thus, aerosol species are indicative of causal microphysical and radiative effects. Different aerosol types may reveal different patterns of ACI. Here, polluted dust (accounting for 34%) and smoke aerosol (accounting for 38%), which are the two main aerosol types occurring in the YRD, are chosen to investigate the variation of cloud parameters with AOD. Smoke (fine absorbing particles) and polluted dust (coarse particles) aerosol are identified using the CALIOP classification. In addition, they have different efficiency for the absorption of sunlight.

Figure 8 shows the variation of cloud parameters with AOD over the YRD, where data points for mixed polluted dust-warm clouds and mixed smoke aerosols-warm clouds are indicated with different colours. Figure 8(a) shows that the CDR is, in general, larger in the presence of smoke aerosol than in the presence of dust. Meanwhile, the cloud fraction is smaller in the presence of smoke, as shown in Figure 8(b). This can be due to the greater efficiency of smoke aerosol particles for the absorption of sunlight than that of dust, resulting in local warming in the presence of smoke aerosol which in turn leads to evaporation of water and thus an increase in small droplets or even complete evaporation of cloud droplets and thus a reduction of cloud cover. Figure 8(c) shows that the cloud optical thickness decreases with increasing AOD for both aerosol types albeit with a low significance as indicated by the small correlation coefficient R. The slope of linear regression of cloud optical thickness against AOD is much stronger in the presence of smoke aerosol than in the presence of dust, indicating that the ACI is stronger for smoke than for polluted dust. In addition to those mentioned, one factor which probably also contributes to the observed difference between effects of smoke and polluted dust is that dust does not absorb sunlight at 0.86μm (Kaufman et al., 2005). Figure 8(d) shows that the slope of linear regression of cloud top pressure against AOD is much stronger for smoke aerosol than that for polluted aerosol, with a correlation coefficient equal to 0.36. Both these results may be due to the higher absorption efficiency of smoke (Small et al., 2011).

3.3.3 Influence of relative humidity on ACI

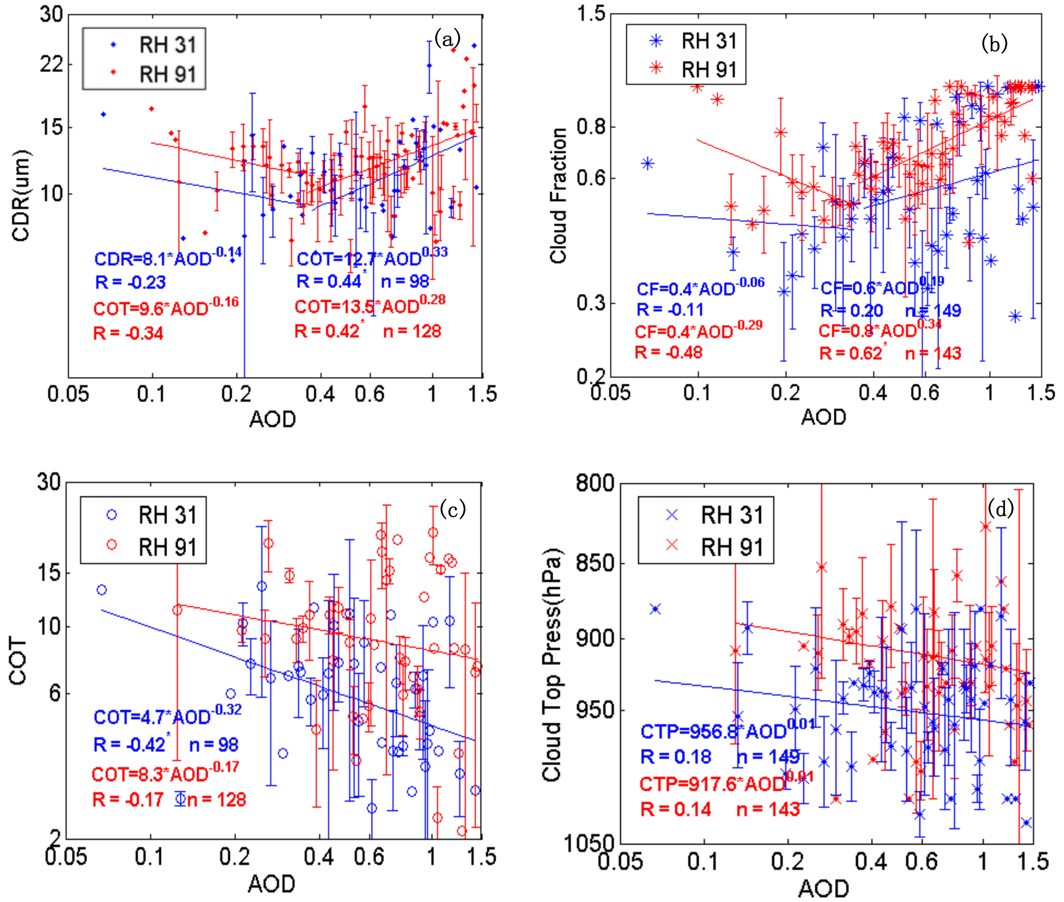

Figure 9. Scatterplots of cloud parameters versus AOD over YRD on log-log scale for cases of low RH (31%) condition (blue) and mixed aerosol-cloud layers under high RH (91%) condition (red), (a) CDR, (b) CF, (c) COT and (d) CTP. The lines present the least squares fits and the resulting relations are presented in each figure. The error bars indicate the statistical uncertainties as in Fig. 6.

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).

Relative humidity (RH) is one of the main factors affecting aerosol particle size and cloud formation. For instance, high RH at cloud base has been reported to affect the relation between aerosol particles and cloud properties (Small et al., 2011). Thus, effects of RH need to be accounted for in aerosol-cloud interaction studies, as reported in the literature (Jeong et al., 2007; Loeb and Manalo-Smith, 2005; Quaas et al., 2010).

The cloud properties versus AOD relationships are classified by RH (at 950hPa) in three equally sized subsets and the mean RH values for each subset are calculated. In figure 9 we show cloud properties as function of AOD for only the lowest RH (31%), representing dry conditions, and the highest RH (91%, above the deliquescence point of ambient particles). Figure 9(a) shows that the CDR is larger in high relative humidity conditions than in low relative humidity conditions, irrespective of the AOD. It is likely that hygroscopic aerosols grow in size caused by condensation of water vapour (Hanel, 1976; Feingold et al., 2003). The increasing RH further increases the probability of the cloud droplet activation and growth of existing cloud droplets as well (Jones et al., 2009). This indicates that high relative humidity conditions can help the formation of larger cloud droplets due to a higher water vapour content in the atmosphere. The cloud fraction is much larger in high relative humidity conditions than in low relative humidity conditions, as shown in Figure 9(b). Figure 9(c) shows that the cloud optical thickness decreases with increasing AOD in both conditions, albeit with a low significance as indicated by the small correlation coefficient R. However, the cloud optical thickness is larger in high relative humidity conditions than in low relative humidity conditions for the entire AOD dataset. In contrast, the cloud top pressure is smaller in high relative humidity conditions than in low relative humidity conditions over the whole range of AOD values (Figure 9(d)). This implies that high relative humidity can promote the formation of thicker and higher clouds.

3.3.4 Influence of boundary layer thermodynamics and dynamics on ACI

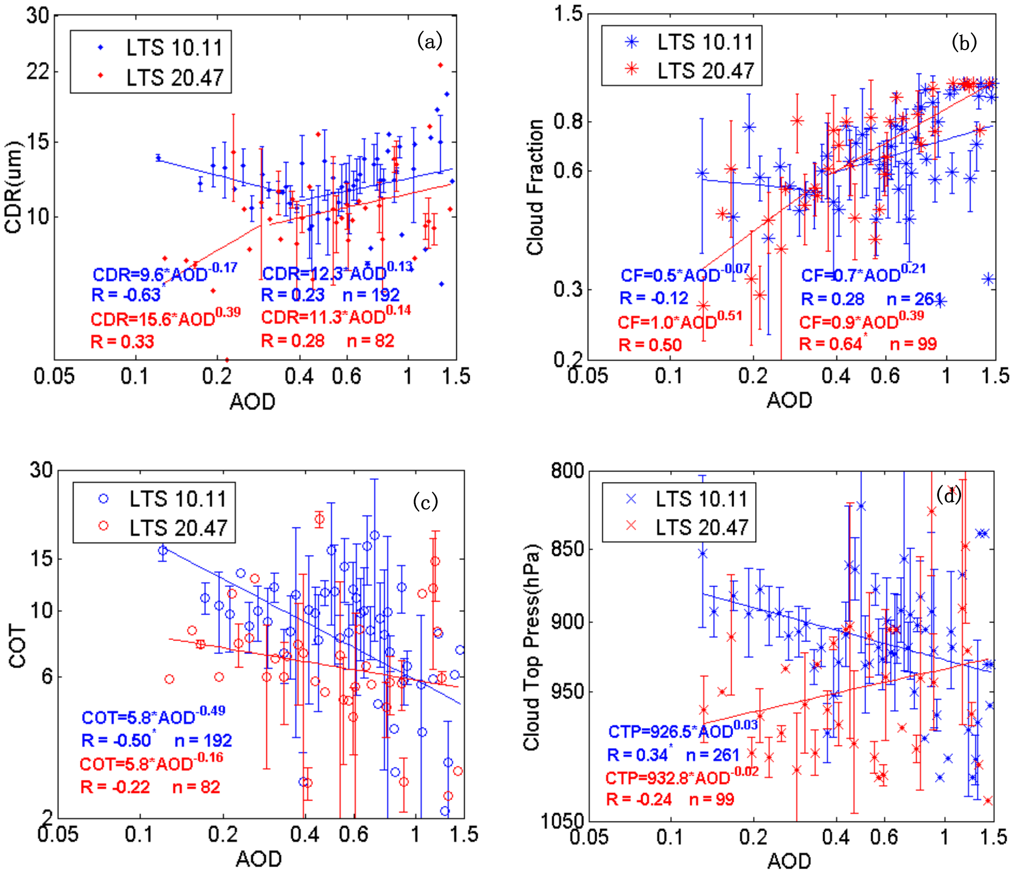

Figure 10. Scatterplots of cloud parameters versus AOD over YRD on log-log scale for cases of low LTS condition (blue) and mixedaerosol-cloud layers under high LTS condition (red), (a) CDR, (b) CF, (c) COT and (d) CTP. The lines present the least squares fits and the resulting relations are presented in each figure. The error bars indicate the statistical uncertainties as in Fig. 6.

The LTS is an indicator for the mixing state of the atmospheric layer adjacent to the surface. It describes to some extent the atmosphere's tendency to promote or suppress vertical motion (Medeiros and Stevens, 2011), which in turn affects cloud properties (Klein and Hartmann, 1993).

Figure 10 shows cloud properties as function of AOD for two different LTS conditions: low LTS, with a mean value equal to 10.11 representing an unstable atmosphere; and high LTS, with a mean value equal to 20.47 representing a stable atmosphere. Figure 10(a) shows that the CDR is larger in unstable atmospheric conditions than in stable conditions, irrespective of the AOD. This indicates that in unstable atmospheric conditions the cloud droplets are larger, which may be due to stronger interaction between aerosols and clouds as a result of better vertical mixing of water vapour. Figure 10(b) shows that the slope of linear regression of cloud fraction against AOD is much stronger for stable atmospheric conditions than for unstable atmospheric conditions in the heavily polluted conditions. This demonstrates that stable atmospheric conditions can promote the formation of a cloud (Small, et al., 2011). A high LTS indicates a strong inversion, which prevents vertical mixing and cloud vertical extent, maintaining a well-mixed and moist boundary layer and providing an environment which favours the development of a low cloud cover. Figure 10(c) shows that the cloud optical thickness is larger in unstable atmospheric conditions than in stable atmospheric conditions. In contrast, the cloud top pressure is smaller in unstable atmospheric conditions than in stable atmospheric conditions for the whole range of AOD values (Figure 9(d)). This indicates that unstable atmospheric conditions can promote the formation of thicker and higher clouds and stable atmospheric conditions can enhance the cloud cover.

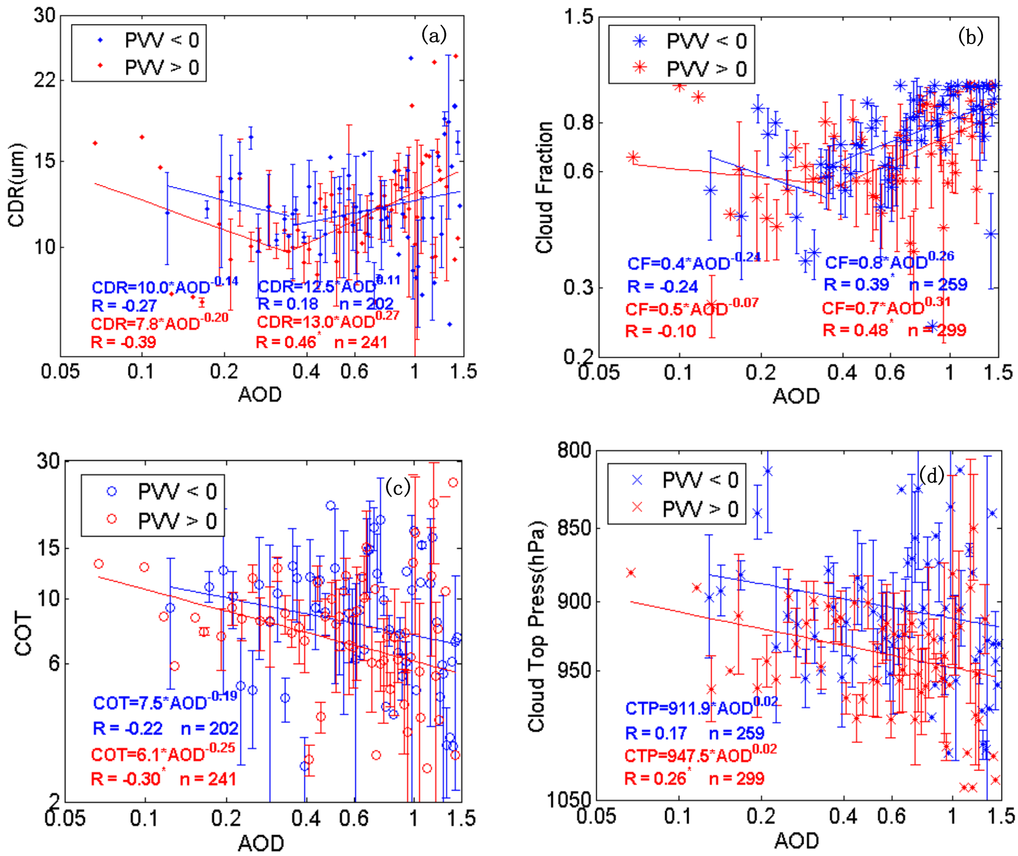

Figure 11. Scatterplots of cloud parameters versus AOD over YRD on log-log scale for cases of PVV<0 condition (blue) and mixed aerosol-cloud layers under high PVV>0 condition (red), (a) CDR, (b) CF, (c) COT and (d) CTP. The lines present the least squares fits and the resulting relations are presented in each figure. The error bars indicate the statistical uncertainties as in Fig. 6.

The PVV, a measure of dynamic convection strength, is very important for cloud formation. In particular, the vertical velocity can be used to determine whether a certain region may be susceptible to cloud development or not. That is, the presence of upward motion, as indicated by negative PVV, can enhance

ACI as it makes the ambient environment favourable for cloud formation, and vice versa (Jones et al., 2009).

Figure 11(a) shows that in moderately polluted condition the CDR is larger in the presence of upward motion of air parcels than for downward motion. This observation indicates that the upward motion of air parcels can promote the formation of larger cloud droplets, thus enhancing ACI. However, the impact of vertical velocity is weak in polluted and heavily polluted conditions. Figure 11(b) shows that the cloud fraction is larger in the presence of upward motion of air parcels than for downward motion of air parcels when AOD is greater than 0.35. This indicates that the upward motion of air parcels can favour cloud development and increase cloud cover in heavily polluted conditions. The phenomenon is not obvious when AOD is smaller than 0.35. These results emphasize the importance of vertical velocity when estimating the potential aerosol effect on cloud droplet effective radius and cloud fraction in polluted conditions. Figure 11(c) shows that the cloud optical thickness is larger in the presence of upward motion of air parcels than for downward motion throughout the range of AOD. In contrast, the cloud top pressure is smaller in the presence of upward motion of air parcels than for downward motion (Figure 9(d)). This implies that upward motion of air parcels can be helpful for the formation of thicker and higher clouds.

3.4 Error sources and uncertainties

Caution is warranted in investigating the satellite-derived relations 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, and dynamic effects (Yuan et al., 2008). Below we discuss several potential factors that may have affected the interaction between aerosols and clouds in our analysis.

Firstly, the correlation between AOD and cloud parameters may be influenced by aerosol size distributions (Small et al., 2011). Since the MODIS retrieval does not provide aerosol size information, it is better to explore the seasonal differences in the observed ACI due to the difference in aerosol emissions between the different seasons. However, the relatively low number of MODIS-CALIPSO coincidences limits the further binning of the data required to investigate this issue. Secondly, what it comes to the occurrence of cloud contamination in the AOD dataset, this is a universal and one of the most difficult problems in aerosol retrieval. Cloud detection is usually not perfect, so that 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 a consistency between aerosol and cloud retrievals from the same instrument which showed 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. Conclusions

The high level of anthropogenic emissions in Eastern China render this area an important hotspot for studying how cloud microphysical properties are affected by anthropogenic aerosols (Ding et al., 2013). Based on the near-simultaneous aerosol and cloud retrievals provided by MODIS, CALIOP and CloudSat, together with the ERA Interim Reanalysis data, we investigated the effect of aerosols, with AOD used as a proxy for the aerosol loading, on micro-physical and macro-physical cloud properties over the Yangtze River Delta for the years 2007 to 2010. In terms of the relative heights of aerosol and cloud layers, well-mixed and separated clouds were defined. A statistical analysis was used to examine the aerosol effects on cloud properties for these two cases. Besides the aerosol impact on CDR, CF, COT and CTP, also the influence of environmental conditions, such as RH, LTS and PVV, on the relation between cloud properties and AOD was studied. In addition, the impact of two different aerosol types, dust and smoke, was explored.

The analysis of the COT-CDR and CWP-CDR relationships for well-mixed clouds indicated that they are affected by the aerosol loading. A statistical analysis of the relation between CWP and COT showed an increase in CWP with an increasing COT, which is in a good agreement with the findings reported by Costantino and Bréon (2013).

Consistent with previous findings, we found that the CDR initially decreases with increasing AOD, followed by an increase after AOD reaches a value of 0.35. This result is consistent with Twomey's hypothesis that increasing aerosol abundance leads to more numerous but smaller cloud droplets at given constant cloud water content. The positive relation between CDR and AOD may be caused by microphysical processes, which is coupled with intense vapour competition and evaporation of smaller droplets as a result of a high abundance of aerosol particles. Also, the analysis of the variation of CF with increasing AOD showed that CF varies with AOD in a way similar to that of CDR. This finding differs from those by Koren et al. (2008) and Small et al. (2011) who observed that an increase in the cloud cover with an increasing AOD, followed by a decrease with higher AOD. COT was found to decrease with an increasing AOD. We argue that the radiative effect and retrieval artefact due to absorbing aerosol might be important factors in determining this relationship. This effect can result in increased cloud evaporation and reduced cloud cover. Meanwhile, CTP tends to increase as aerosol abundance increases, indicating that the aerosol is prone to expand the horizontal extension. In other words, we found that for well-mixed clouds over the YRD, the CDR becomes smaller with the increase of AOD in moderately polluted conditions which in principles in line with the Twomey effect, yet, the cloud fraction indicates a weak decrease which could be attributed only to the weak influence of evaporation caused by absorption of aerosols.

On the other hand, in polluted and heavily polluted conditions, a reduced cloud coverage can result in more solar radiation reaching the surface, causing surface heating and thus raises the surface temperature, which then destabilizes the atmosphere. The resulting advection transports water vapour from the surface to higher levels in the atmosphere, therefore producing more cloud. Meanwhile, CDR becomes larger as a result of the stronger water vapour competition in polluted and heavily polluted conditions. The COT decreases with the increasing values of AOD throughout the AOD range due to the radiative effect and possible retrieval artefacts. The behaviour of CTP is consistent with that of

COT, with the cloud getting thinner but with larger cover, so that CTP becomes larger with an increasing AOD.

Furthermore, joint correlative analysis of different aerosol and cloud properties revealed that smoke aerosols have a stronger impact on aerosol-cloud interaction due to their stronger absorption of solar radiation compared with polluted dust. Therefore, we can conclude that absorbing aerosols play an important role in the aerosol cloud interaction.

Constrained by relative humidity and boundary thermodynamic and dynamic conditions, the variation of cloud properties in response to aerosol abundance was analysed. In general, a high relative humidity can promote the formation of larger cloud droplets and expand cloud formation, irrespective of the vertical or horizontal level. With regard to LTS, stable atmospheric conditions can enhance the cloud cover horizontally. However, unstable atmospheric conditions can be helpful for the formation of thicker and higher clouds. Dynamically, an upward motion of air parcels can also facilitate the formation of thicker and higher clouds. Besides the meteorological controls mentioned above, other factors may be important in generating relations between aerosol and cloud properties, such as temperature advection. These results suggest that effects of ambient meteorological environments need to be considered when exploring the aerosol indirect effect. In summary, this study will greatly help us to understand the mechanisms of aerosol-cloud interaction and ultimately of aerosol indirect effects over the YRD.

Acknowledgements

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