Satellite-based estimate of the variability of warm cloud properties associated with aerosol and meteorological conditions

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Abstract. Aerosol-cloud interaction is examined using ~~four~~ ten years of data from the MODIS/Terra (morning orbit) and MODIS/Aqua (afternoon orbit) satellites. Aerosol optical depth (AOD) and cloud properties retrieved from both sensors are used to explore in a statistical sense the morning-to-afternoon variation of cloud properties in conditions with low and high AOD, over both land and ocean. ~~The results show that the morning-to-afternoon variation of cloud properties during the 3 hours between the Terra and Aqua overpasses have similar patterns (increase or decrease) over land under both low and high AOD conditions~~ The results show that the interaction between aerosol particles and clouds is more complex and of greater uncertainty over land than over ocean. The variation in d(Cloud_X), defined as the mean change in cloud property Cloud_X between the morning and afternoon overpasses in high AOD conditions minus that in low AOD conditions, is different over land and ocean. This applies to cloud droplet effective radius (CDR), cloud fraction (CF) and cloud top pressure (CTP), but not to cloud optical thickness (COT) and cloud liquid water path (CWP). Both COT and CWP increase over land and ocean after the timestep, irrespective of the AOD. However, the initial AOD conditions can affect the amplitude of variation of COT and CWP. The effects of initial cloud fraction and meteorological conditions on the change in CF under low and high AOD conditions after the 3 hours timestep over land are also explored.~~,~~ ~~There are~~ T~~t~~wo cases are considered: (1) when the cloud cover increases; (2) when the cloud cover decreases. ~~From~~ For both ~~two~~ cases, we find that almost all ~~the~~ values of d(CF) are positive, indicating that the variations of CF are larger in high AOD than that in low AOD after the 3 hours timestep. The results also show~~ing~~ that large increase of cloud fraction occurs when scenes experience large AOD and stronger upward motion of air parcels. Furthermore, the increase rate of cloud cover is larger for high AOD with increasing RH~~scenes with large cloud fraction experience large AOD and larger RH~~ when RH ~~I~~is larger than 20%. We also find that smaller increase of cloud fraction occurs when scenes experience larger AOD and larger initial cloud cover. ~~upward motion of air parcels can enhance the cloud cover much more when AOD is high than when it is low. In contrast, the increase of cloud cover~~of ~~with increasing relative humidity is much stronger in a relatively clean atmosphere with low AOD than in a more polluted atmosphere. Meanwhile, stable atmospheric conditions favour the development of a low cloud cover, especially when AOD is high.~~ Overall, the analysis of the diurnal variation of cloud properties provides a better understanding of aerosol-cloud interaction over land and ocean.

Key words: MODIS, cloud development, aerosol-cloud interaction, urban clusters, ocean

1 Introduction

Clouds and cloud systems are crucial elements in the energy cycle of our planet (Hartmann et al., 1992; Webb et al., 2006). Clouds affect the global energy budget by reflecting incoming solar radiation, and thus cool the Earth surface, and by absorption and re-emitting outgoing terrestrial radiation which contributes to warming of the surface. In addition to the radiative effects, clouds also influence the hydrological cycle of the Earth through precipitation (Stephens et al., 2002). Due to interactions with aerosols, the climatic effects of clouds are further complicated (Rosenfeld, 2000; Twomey, ~~2007~~1974; Twomey, 1977). Aerosols can serve as cloud condensation nuclei (CCN), depending on their hygroscopic properties, and when activated they can change the cloud microphysical properties. The increase of CCN, while the liquid water path remains constant, usually results in more numerous cloud droplets with smaller cloud droplet radius (CDR) due to the competition for the same amount of water vapour. Thus, cloud albedo increases and the smaller cloud droplet effective radius in most cases results in the suppression of precipitation~~ in most cases~~, which in turn results in a longer cloud lifetime, and maintaining a larger liquid water path (Albrecht, 1989; Feingold et al., 2001). Therefore, it is important to understand the interaction between aerosols and clouds and the effect of different processes on cloud development.

Numerous studies have shown that aerosol particles can affect cloud properties on regional and global scales (Krüger and Graßl, 2002; Menon et al., 2008; Rosenfeld et al., 2014; Sporre et al., 2014; Saponaro et al., 2017). Satellite measurements suggest that the cloud droplet effective radius (CDR) decreases with increasing aerosol optical depth (AOD, which is used in this paper as a proxy for aerosol concentration), which is consistent with Twomey's theory (Kaufman et al., 2005; Matheson et al., 2005; Meskhidze and Nenes, 2010). However, other observational and model studies reported that CDR tends to increase with aerosol loading in some study areas, especially over land (Feingold et al., 2001; Yuan et al., 2008; Grandey and Stier, 2010; Liu et al., 2017). A different behaviour of cloud cover as a function of AOD for different aerosol loadings (low or high) has been found by Kaufman and Koren (2006) and Koren et al. (2008). However, the observed correlations between aerosol and cloud cannot be simply attributed to the effects of aerosols on clouds alone since other factors such as variations in meteorological conditions could play a role (Loeb and Schuster, 2008; Reutter et al., 2009; Koren et al., 2010; Su et al., 2010; Stathopoulos et al., 2017). "Snapshot" studies, where the aerosol and cloud properties are retrieved at the same time, have the advantage that they represent the total time-integrated effect of aerosols on cloud properties (Meskhidze et al., 2009; Gryspeerdt et al., 2014). However, the use of "snapshot" correlations is limited to a single overpass time and limits the ability to distinguish aerosol-cloud interactions from meteorological covariation or retrieval errors (Gryspeerdt et al., 2014). Therefore, the history of meteorological forcing is an important determinant of cloud state. Matsui et al. (2006) investigated the properties of low clouds derived from semiglobal observations by the Tropical Rainfall Measurement Mission (TRMM) and explored the correlations of these cloud properties with aerosols (as indicated by the aerosol index or AI) and with lower-tropospheric stability (LTS) on a diurnal scale. They found that aerosols affect the CDR stronger for low LTS than for high LTS. Mauger and Norris (2007) used MODIS/Terra data to examine the evolution of marine boundary layer clouds over several days but they may have missed important effects occurring on a sub-daily timescale. Meskhidze et al. (2009) investigated the evolution of cloud properties between the MODIS/Terra and MODIS/Aqua overpasses as a function of MODIS/Terra AOD and found an apparent increase in the breakup rate of stratocumulus clouds in high AOD environments. However, they did not explain meteorological covariation that may generate spurious correlations.

Considering the complex aerosol composition and increasing aerosol trend during the last decades over eastern China (Guo et al., 2011), a systematic assessment of the effect of aerosols on the properties of warm clouds is ~~desperately~~ needed, over both land and ocean. In this paper, aerosol-cloud interaction is examined using multi-year statistics of remotely sensed data from the two MODIS sensors aboard NASA's Terra (daytime equator crossing time at 10:30 LT) and Aqua (daytime equator crossing time at 13:30 LT) satellites. The retrieval of the AOD and cloud properties from both sensors allows us to explore the morning-to-afternoon variation of cloud properties in conditions with either low or high AOD, over land and over ocean, and for different climate regimes. This variety of conditions allows us to identify similarities and differences in the effects of aerosols on clouds and thus better understand aerosol-cloud interaction. We also explore the effect of meteorological history on the interaction between aerosols and clouds. ~~In this paper, w~~We focus on low-level water clouds. The paper is organized as follows. The data and region of interest are described in Section 2. The main methodology is introduced in ~~section~~ Section 3. The results and analysis are presented in ~~section~~ Section 4. Overall conclusions and potential future improvements are discussed in ~~section~~ Section 5.

2 Approach

2.1 Study area

Aerosol concentrations in Eastern China are very high due to both direct emissions and secondary aerosol formation from precursor gases such as NO_2, SO_2 and VOCs. They are produced by anthropogenic activities such as industry, transportation and heating, black carbon and other carbonaceous aerosols produced by biomass burning, dust aerosols produced from the deserts, etc. Aerosol particles influence the local climate such as monsoon intensity and the distribution of precipitation.~~, conversely, monsoon i~~In eastern China, the monsoon in turn ~~also~~ plays an important role in the wet deposition and transport of aerosol particles (Li et al., 2016). The Asian monsoon system plays an important role in the precipitation across the country (Kourtidis et al., 2015). In early April, the pre-monsoonal rain period starts over southern China and the summer monsoon rain belt moves northward to the Yangtze River basin in June. Further, the rain belt arrives in northern China in July and the monsoon rain belt propagates back to southern China in August. The length of the rain season differs between southern and northern China with the migration of the monsoon across China (Song et al., 2011). Based on these characteristics, four regions with different aerosol emission levels and climate characteristics were selected to study the indirect effects of aerosol particles on cloud micro- and macro-physical properties. The Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD) and Pearl River Delta (PRD) urban clusters are characterized as a temperate monsoon climate region, a subtropical monsoon climate region, and a tropical monsoon climate region, respectively. The BTH domain (35.5°N-40.5°N, 113.5°E-120.5°E) is an area with high AOD levels due to rapid industrial and economic development (Fig. 1). The YRD domain (28°N-33°N, 117°E-122°E) is a major source region of black carbon (Streets et al., 2001; Bond et al., 2004) and sulfate (Lu et al., 2010). The PRD domain (21.5°N-24.5°N, 111.5°E-115.5°E) is an area within the intertropical convergence zone (ITCZ) migration belt, with high anthropogenic aerosol emissions (Streets et al., 2003; Streets et al., 2008; Lei et al., 2011). In addition, one domain (20°N-25°N and 125°E-130°E), which is located in the Eastern China Sea (ECS for short), has been selected as study area for comparison. The ECS domain is relatively clean, but it is often impacted by aerosol particles transported from the highly industrialized eastern China (Wang et al., 2014). The study period is 10~~4~~ years, i.e. 2008-~~2011~~2017.

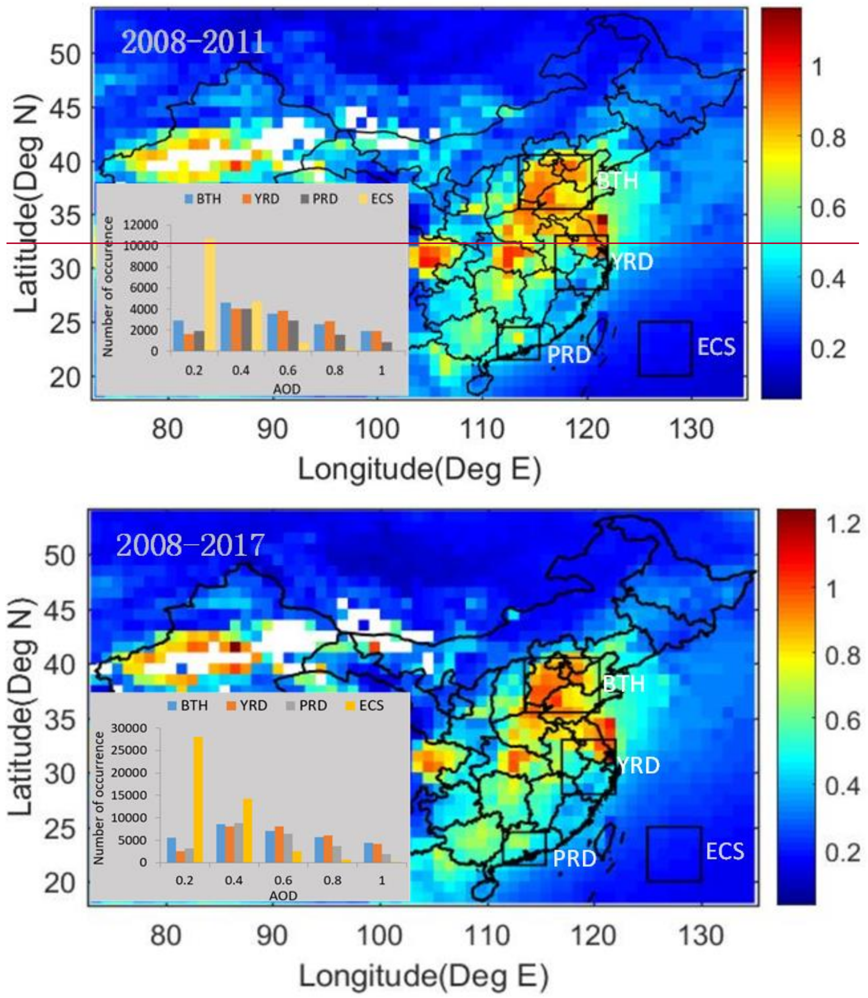


Figure 1. Map of MODIS/AQUA level 3 AOD over Eastern China averaged over the period from 2008 to ~~2011~~2017. The location of the four clusters (three urban and one ocean) studied here (Beijing-Tianjin-Hebei: BTH, Yangtze River Delta: YRD, Pearl River Delta: PRD and Eastern China Sea: ECS) are marked with black rectangles. The inset shows a histogram for the occurrence of AOD values in each of the four clusters during the period 2008-~~2011~~2017.

2.2 Data used

The aerosol and cloud properties used in this study were derived from the MODIS instruments on the Terra and Aqua satellites. Since these instruments are of the same design, errors due to instrument differences are minimal although some differences have been reported due to degradation of MODIS/Terra (Xiong et al., 2008; Levy et al., 2010~~; Xiong et al., 2008~~). The MODIS L3 collection ~~5.1~~6.1 data (which was downloaded from https://ladsweb.modaps.eosdis.nasa.gov/) provides daily aerosol and cloud parameters on a 1° by 1° spatial grid. The time difference between the Terra and Aqua overpasses is about three hours, with variations due to swath width. In the following, the time difference between the MODIS/Terra and Aqua observations is referred to as the timestep. The application of daily MODIS satellite data on a 1° by 1° spatial grid in this study on aerosol-cloud interaction (ACI) ensures that the aerosol and cloud retrievals are coincident. The MODIS instruments have 36 spectral bands, the first seven of these (0.47- 2 .1 3 μ m) are used for the retrieval of aerosol properties (Remer et al., 2005) while cloud properties are retrieved using additional wavelengths in other parts of the spectrum (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. In this study on ACI we use the AOD at 550 nm (referred to as AOD throughout this manuscript), CDR, cloud liquid water path (CWP), cloud optical thickness (COT), cloud fraction (CF), cloud top pressure (CTP) and cloud top temperature (CTT) from both instruments. AOD is used as a proxy for the amount of aerosol particles in the atmospheric column to investigate ACI (Andreae, 2009; Kourtidis et al., 2015). To reduce a possible over-estimation of the AOD, cases with AOD greater than 0.8 were excluded from further analysis. The focus of this study is on warm clouds with CTP ~~greater~~ larger than 700 hPa, CTT ~~greater~~ larger than 273K and CWP lower than 200 g m^{-2}, as most aerosols exist in the lower troposphere (Michibata et al., 2014).

In addition, to explore the effect of meteorological conditions on ACI, we use the daily temperature at the 1000 hPa and 700 hPa levels, relative humidity at the 750hPa level and pressure vertical velocity (PVV) at the 750 hPa level. LTS is defined as the difference in potential temperature between the free troposphere (700hpa) and the surface, which can be regarded as a measure of the strength of the inversion that caps the planetary boundary layer~~is representative of typical thermodynamic conditions~~ (Klein and Hartmann, 1993; Wood and Bretherton, 2006). These meteorological data were obtained from daily ERA Interim Reanalysis data which contains global meteorological conditions on a grid of 1°×1° with 37 levels in the vertical (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 meteorological properties were resampled to 10:30 (local time) by taking a weighted average of the properties at the two closest times (00:00 UTC and 06:00 UTC) provided by ERA Interim.

In this study, high and low AOD are defined as the highest and lowest quartile for each 1°×1° location to reduce climatological spatial gradients in aerosol and cloud parameters. As a result, the difference between high and low AOD varies by location. So, for each 1° x 1° grid cell, ~~1457~~ 3642 data samples are available for the 10~~4~~-year study period.

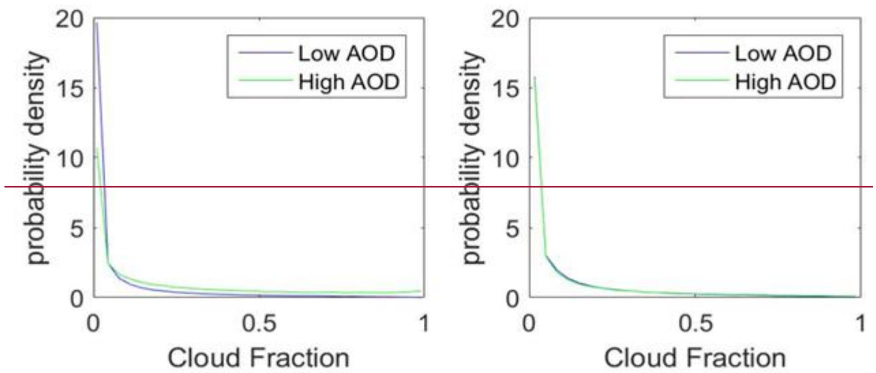
3 Method

3.1 Normalization for initial background

For the comparison of the difference in cloud properties in high and in low AOD conditions and the change in this difference during the time step, we need to ensure that the initial conditions are similar, i.e. the probability distributions of a cloud parameter Cloud_X at the start of the time step for the low and high AOD cases should be similar. Any change in this distribution at the end of the time step can then be attributed to changes in cloud properties due to aerosol and/or meteorological effects. To reduce the difference between the initial probability distribution of Cloud_X in high and low AOD conditions at the start of the timestep, normalized histograms of cloud properties and meteorological parameters are made for high and low AOD conditions following the method described by Gryspeerdt et al. (2014).

~~It is necessary to reduce possible non-aerosol effects linking cloud properties and AOD at the start time to reveal the strong link between cloud properties and AOD.~~ In Fig. 2 we illustrate the process to remove possible effects linking, as an example, CF and AOD. Normalized histograms of CF are made for the high and low AOD conditions following Gryspeerdt et al. (2014) with the difference that in the current study AOD is used instead of AI (Andreae, 2009; Kourtidis, et al., 2015). The CF probability density functions for low and high AOD conditions at the start time are different as illustrated ~~for CF~~ in Fig. 2a. This difference indicates a link between CF and AOD at the start of the time step which needs to be removed to detect the effect of changes during the time step. This is achieved, following the process described in more detail by Gryspeerdt et al. (2014). In brief, for each bin data points are drawn out randomly from the conditions with the larger probability density frequency until both distributions match. This is performed independently for each bin and the entire process is repeated until the ~~conditions have sufficiently similar~~ normalised histograms in both AOD conditions are similar. As a result of this normalization process, the ~~After that, both conditions have almost the same~~ CF distributions at the start of the timestep are nearly identical for both AOD conditions~~through the normalization process~~, i.e. ~~indicating that~~ the non-aerosol effect linking CF and AOD has been removed. This technique has also been applied to ensure that the high and low AOD conditions have the same probability distributions ~~of~~ for CDR, COT, CWP and CTP at the start time. Among those cloud properties, this process of ~~, even though we know that the~~ normalization ~~for the cloud fraction~~ has the greatest effect on the cloud fraction and it~~'~~s dependence on ~~-on the relationship between~~ aerosol~~-and~~ -cloud interaction~~among those cloud properties~~~~even though we find that the normalization for the cloud fraction made the biggest difference by far~~. ~~In the further analysis~~Throughout the work, we only take a subset of original data by removing random samples until the histograms are similar.

Note that here and in the following sections, normalised histograms of cloud properties ~~are made~~ for the high and low AOD populations ~~based~~ are made ~~on~~for the whole region (Section 3.1), because ~~as there is relatively small~~ the data volume based on each 1° x~~by~~ 1° location is relatively small. However, ~~T~~the difference between~~of~~ the cloud properties ~~between~~for ~~the~~ low and high AOD at the start time is based on each 1° x 1° location (Section 4.1). So the difference of the cloud properties between the low and high AOD at the start time is not zero.



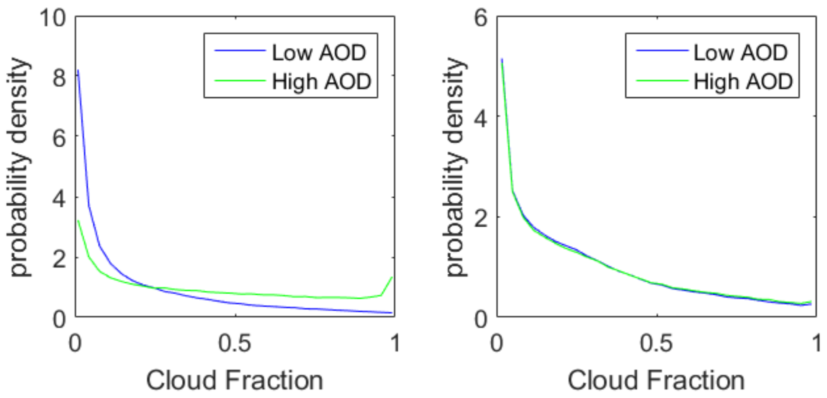


Figure 2 An example of the p~~P~~robability density distribution of warm cloud fraction (CF) for low and high AOD conditions. (a) there is a strong link between AOD and CF before histogram normalization, (b) the link is reduced after histogram normalization.

3.2 The definition of d(Cloud_X)

After removal of the potential relationships between AOD and cloud parameters at the time of the Terra (morning) overpass, as described in Sect. 3.1, effects of aerosol particles on cloud properties are investigated from the ~~recovery~~ change in~~of~~ the relationship between AOD and cloud parameters over the timestep. For cloud property Cloud_X (where X = CF, COT, CWP, CDR or CTP), the change during the timestep is indicated by ΔCloud_X. The mean ΔCloud_X for high AOD is then indicated by $\overline{\Delta\text{Cloud\_X[High AOD]}}$ ~~ΔCloud_X[High AOD]~~ and similar for low AOD. The difference between the mean change in Cloud_X during the timestep in high and low AOD conditions is then indicated by d(Cloud_X):

$$d(\text{Cloud\_X}) = \overline{\Delta\text{Cloud\_X[High AOD]}} - \overline{\Delta\text{Cloud\_X[Low AOD]}}$$

~~For example, d(CWP) would be the difference between the mean change in CWP in high AOD conditions minus that in low AOD conditions.~~

The high AOD is representative of polluted atmospheric conditions, and the low AOD is representative of clean atmospheric conditions. The difference (d(Cloud_X)) between the mean values of the cloud property Cloud_X during clean (low AOD) and polluted (high AOD) conditions indicates the effect of these two aerosol cases on the cloud property Cloud_X. For example, d(CWP) would be the difference between the mean change in CWP in high AOD conditions minus that in low AOD conditions.

Student's t test is used to determine whether two data sets ~~of data~~ are significantly different from each other. The marker ** at the top right corner of symbol "+" (or "-") denotes that the difference between a change in cloud property and zero is significant (at 95% confidence level).

4 Results and Discussion~~s~~

4.1 The difference of cloud properties between the low and high AOD at the start time

The difference in the mean cloud properties (CDR, CF, COT, CWP and CTP) during high and low AOD conditions at the start time for each 1°×1° grid cell, i.e., $\left\{\overline{\text{Cloud\_X[High AOD]}} - \overline{\text{Cloud\_X[Low AOD]}}\right\}\Big|_{t=0}$ represents the change in cloud properties due to the higher AOD. Figure 3 shows the spatial distributions of these differences (left column) and sample series of the difference (right column) for the four regions of interest~~ (right column)~~. The selection of samples for each region is according to the pixels in the region. ~~There are large variations in the response of the cloud parameters to the higher AOD in the four regions.~~ Figures 3(a1-a2) show that over the ECS, CDR is smaller at high AOD than at low AOD, which is consistent with Twomey's effect. In contrast, over the ~~BTH and the YRD~~three urban clusters, CDR is larger at high AOD. This behavior has been observed before for warm clouds in conditions with high AOD (Liu et al., 2017) and may result from the intense competition for the available water vapour and the evaporation of smaller droplets as a consequence of the high aerosol abundance over these regions (Yuan et al., 2008; Tang et al., 2014; Wang et al., 2014; Liu et al., 2017). ~~However, over the PRD urban cluster no statistically significant difference in CDR is observed between high and low AOD.~~ For COT (figures 3(b1-b2)) the values are significantly higher at high AOD over the ECS and the BTH, however, COT does not show a significant difference between the situations at low and high AOD over the YRD and PRD. These results indicate that there is no clear dependence of COT on aerosol load, and also the aerosol type may influence the aerosol effect on COT. ~~and the PRD, in contrast to the smaller value observed over the BTH and the YRD urban clusters. Likely, this is due to the radiative effect and possible retrieval artefacts as explained in Liu et al. (2017). They inferred that the evaporation of cloud droplets caused by locally absorbing aerosol makes clouds thinner and the presence of absorbing aerosol may reduce the satellite-retrieved COT.~~ Figures 3(c1-c2) show~~s~~ that CWP is lower at high AOD over the ECS, which is in clear contrast with the so-called "lifetime effect" proposed by Albrecht in 1989~~not in agreement with COT variation~~. In contrast, over the BTH~~PRD~~, CWP behaves similar to COT and is higher at high AOD. Furthermore, CWP is also higher at high AOD. Ackerman et al. (2004) reported that CWP is not generally observed larger, but significantly smaller in high AOD conditions. They reported that CWP response to the increasing AOD is determined by the balance of two competitive factors: moistening from precipitation decrease and drying from increasing entrainment of dry overlaying air. ~~However, over the BTH and the YRD urban clusters CWP does not show a significant difference between the situations at low and high AOD. Over the ECS, the change in CF between low and high AOD is similar to that of CTP (Figs. 3(d1-d2, e1-e2)): both parameters are larger at high than at low AOD over that area.~~ With increasing AOD, CF does not show any significant correlation between changes in AOD and CDR variations over the BTH and YRD. However, CF is larger at high AOD over the PRD and ECS. Wang et al., (2014) also found that when aerosol loading is relatively small, cloud cover is found to increase over the YRD and ECS in response to aerosol enhancement regardless of RH conditions. Meanwhile, over the YRD urban cluster ~~CF is lower and~~ CTP is higher at high AOD, as suggested by Liu et al. (2017). In contrast, CTP is lower at high AOD over the BTH and ECS. Many studies have also reported that with higher cloud altitude CTP decreases~~d~~ in most of the places as AOD increases~~d~~ except for some regions at low AOD (Myhre et al., 2007; Kaufman et al., 2005 and Alam et al., 2010). This might have resulted from the suppression of ~~the~~ precipitation by increasing cloud lifetime and thus also affecting the cloud albedo and ~~changing the~~ cloud top pressure. ~~and the PRD urban clusters CF is higher and CTP is lower at high AOD. Overall, it looks like the aerosol-cloud interactions behave~~

~~quite similar, over the BTH and YRD urban clusters, both of which have high pollution levels, while over the PRD they show the opposite behavior. The difference in the response of the cloud parameters to low and high aerosol conditions over the four regions may be caused by the difference in pollution levels or pollution types including black carbon, sulphate, sea spray, etc.~~

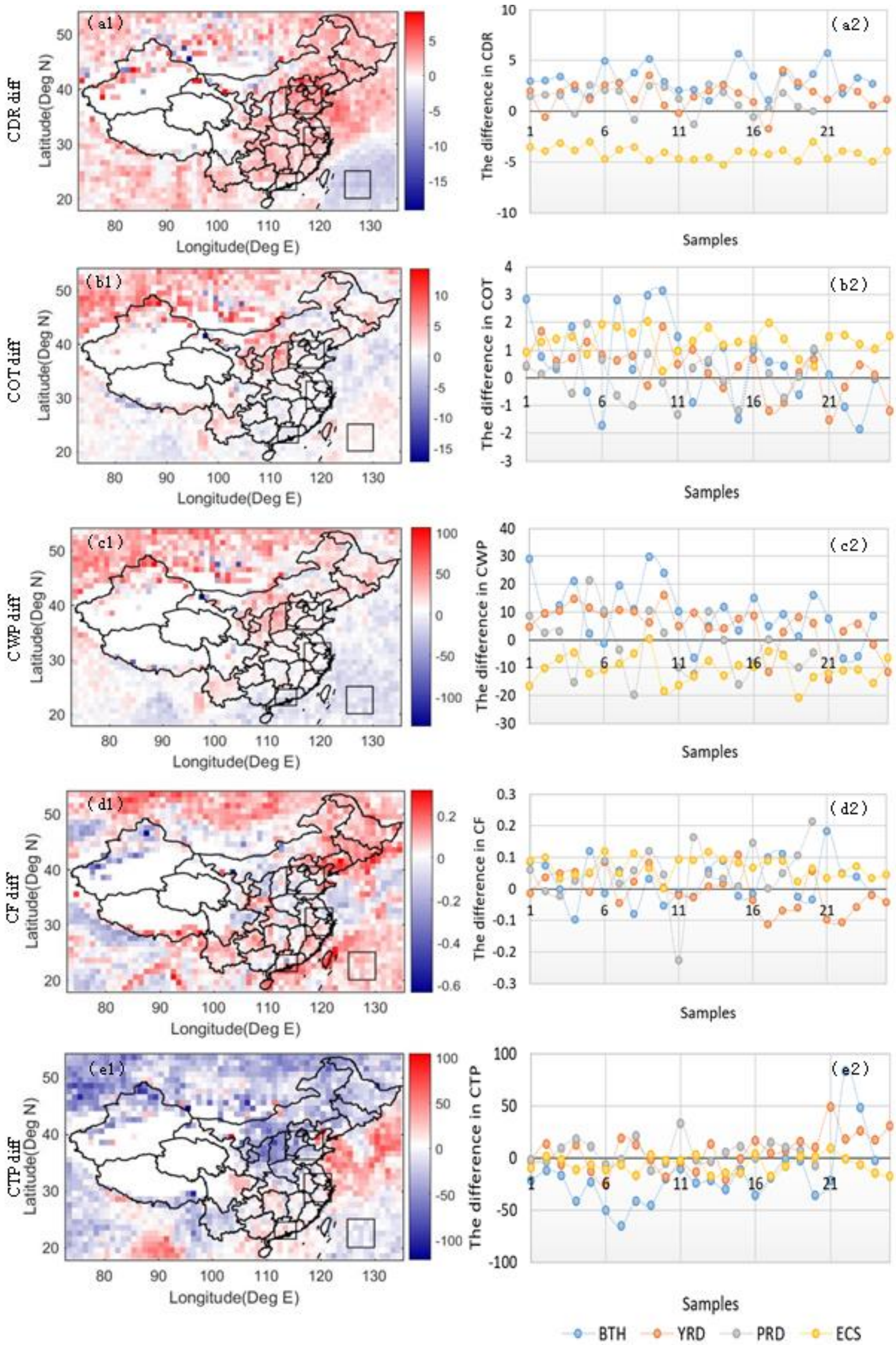


Figure 3 Spatial distribution of the differences in cloud properties (top to bottom: CDR, COT, CWP, CF and CTP) between the highest and the lowest MODIS AOD quartiles (highest - lowest) at the start time of the timestep (MODIS/Terra) (left, a1-e1) and sample series of the differences in cloud properties (CDR, COT, CWP, CF and CTP) between the highest and the lowest MODIS AOD quartiles (highest - lowest) at the start time of the timestep (MODIS/Terra) (right, a2-e2) over Eastern China for the time period 2008-~~2011~~2017. See legend at the bottom for the meaning of the colours identifying the different regions.

To better characterize the variation in cloud properties between high and low AOD, Table 1 summarizes the difference ~~of~~in cloud properties between high and low AOD at start time~~the responses of cloud properties to the increasing AOD at the start time~~ for the four study areas. We find that different regions with various aerosol emission levels and different climate characteristics show different ACI patterns. Some links between aerosol and cloud in the four regions are different from those of previous studies over China (Wang et al., 2014; Tang et al., 2014; Kourtidis et al., 2015; Liu et al., 2017), itwhich might be due to the use of different data sets (MODIS C6.1 versus older versions), hypothesis and target areas characterized by complex aerosol compositions and varying meteorological conditions. Over the ECS region, CDR and CWP in high AOD conditions are smaller than at low AOD, but COT, CF and CTP are higher. Over the BTH urban cluster, the higher AOD results in higher CDR and CF, while COT and CTP are smaller. Over the YRD urban cluster, CDR and CTP are larger, but COT and CF are smaller at high AOD. Over the PRD urban cluster, COT, CWP and CF are larger, but CTP is smaller at high AOD. Overall, the result implies that the interaction between aerosol particles and clouds is more complex and less predictableof greater uncertainty over land (BTH, YRD and PRD) than over ocean (ECS). Jin and Shepherd (2008) also noted that aerosols affect clouds more significantly over ocean than over land. They suggested that dynamic processes related to factors like urban land cover may play at least an equally critical role in cloud formation.

Table 1 The responses of cloud properties to the increasing AOD

Parameters	AOD	CDR	COT	CWP	CF	CTP
BTH	+	+**	+**	+**	+	-**
YRD	+	+**	+	+**	-	+**
PRD	+	+**	+	-	+**	+**
ECS	+	-**	+**	-**	+**	-**

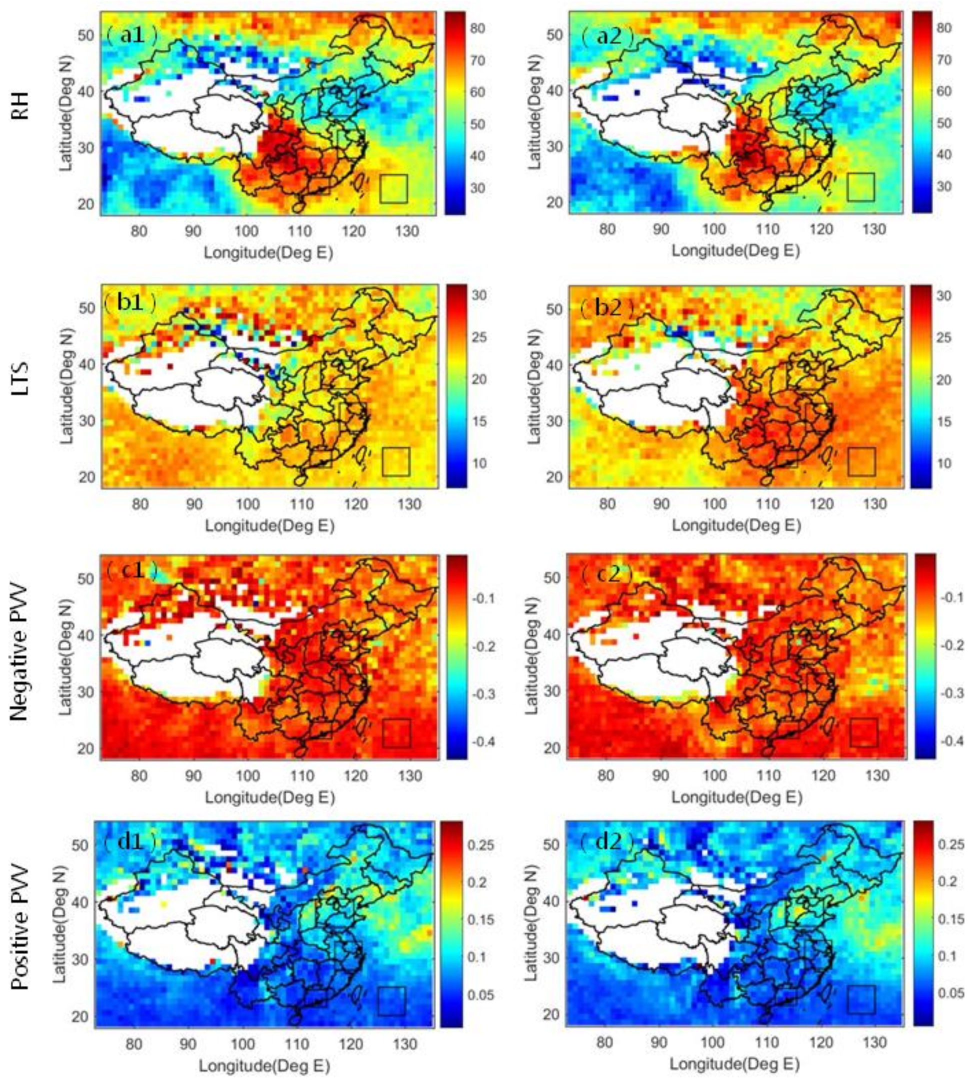
Note：'+' indicates increasing, '-' indicates decreasing and ** at the top right corner of the symbol "+" (or "-") denotes that the difference between a change in cloud property and zero is significant (at 95% confidence level).

Parameters	AOD	CDR	COT	CWP	CF	CTP
BTH	+	+	-	~	+	-
YRD	+	+	-	~	-	+
PRD	+	~	+	+	+	-
ECS	+	-	+	-	+	+

Note：'+' indicates increasing, '-' indicates decreasing and '~' indicates the response of cloud properties to the increasing AOD is not significant.

4.2 The meteorology of the four target regions

The meteorological and aerosol effects on clouds are reported to be so tightly connected at the same time, so itand this connection -must be accounted for in any study intoof aerosol-cloud interactions (Stevens and Feingold, 2009; Koren et al., 2010a). Even Although normalized histograms of meteorological parameters are made for high and low AOD conditions at the start time, but it isthe normalization described in Sect. 3.1 is based on the whole region. So the dDifferences inof meteorological conditions may still existoccur between each 1° by 1° grid cell. In this study, we analyze the meteorology of the different regions, helpingin support of the interpretation of the regional variation of the relationships between aerosols and clouds.



~~Figure 4 Spatial distributions of meteorological parameters (top to bottom: RH, LTS, positive PVV and negative PVV) at the start time of the timestep (MODIS/Terra) for low AOD populations~~conditions (left, a1-d1) and for high AOD ~~populations~~conditions (right, a2-d2). All the data are averaged over all years between 2008 and 2017.

The spatial variations of the aerosol and cloud properties over the four regions, averaged over the years 2008-2017, are shown in Fig. 4. Over the urban clusters, we can see an increasing north–south pattern in RH and LTS, with the lowest values found in the PRD. For the negative PVV, the spatial distributions for the low and high AOD situations ~~of the PVV~~ are remarkably similar ~~to each other~~, with the highest values over the BTH and ~~and~~ decreasing toward the south to near zero over the PRD. ~~However~~In contrast, ~~for~~ the positive PVV~~, the PVV~~ is ~~low~~smallest over the BTH, with little variation over the study area. Overall, the ~~spatial distribution of~~ meteorological parameters over the YRD and PRD are similar to those over~~of~~ the ECS, irrespective of the AOD. Furthermore, the LTS is significantly larger in the high AOD conditions for all ~~the~~ four regions. Zhao et al. (2006) proposed that the enhancement in ~~the~~ atmospheric stability tends to depress upward motion and precipitation, leading to an increase in aerosol particles. The spatial distributions of both positive and negative PVV in the low AOD conditions are similar to those in ~~the~~ high AOD conditions.

4.3~~2~~ The mean change in cloud properties over the timestep for low and high AOD

The differences between the mean afternoon and morning values of cloud properties in each 1°×1° grid cell in either low or high AOD conditions in each 1°×1° grid cell minus those in the morning for low/high AOD shows the variation of cloud properties during 3 hours of cloud evolution at low/high aerosol concentrations. Figure 4 5 presents the spatial distributions (left, a1-e1) and the sample series (right, a2-e2) of differences in cloud properties (CDR, COT, CWP, CF and CTP) after this 3-hour period for the lowest MODIS/Terra AOD quartiles. Figure 65 shows the spatial distributions (left, a1-e1) and sample series (right, a2-e2) of these differences for the highest MODIS/Terra AOD quartiles.

Overall, we look at statistics for a large dataset of 104 years. Concerning the effect of aerosol loading on cloud parameters in each urban cluster, a decrease of CF occurs over the BTH for low AOD conditions, which is opposite to the CTP variation for both two AOD conditions.none of the cloud parameters show a significant increase or decrease over the BTH under either low AOD or high AOD conditions, respectively. For the variations of CDR and CF over the YRD urban cluster, a significant increase occurs under high AOD conditions, which may be attributed to the higher RH (see figure 4(a1, a2)). And fAs regardsor the variation of CF and CTP, a significant decrease occurs under low AOD conditions. but for the variation of CTP, a significant decrease occurs over the YRD for both low and high AOD conditions. By contrast Likewise, an increases of the CDR, COT and CWP were was observed for both low and high AOD conditions over the PRD urban cluster., which is statistically significant. Furthermore, decreases of CF and CTP were observed for low AOD conditions and increases of CF and CTP were observed for high AOD conditions. From the perspective of considering all urban clusters (BTH, YRD and PRD), both COT and CWPall studied cloud properties (CDR, COT, CWP and CF) increase except CTP over land afterduring the 3 hours timestep, which decreases during the timestep, for both low and high AOD (see red samples plot in Figures 4 and 5). Overall, the variation in cloud properties after the timestep over BTH is less significant than those over the YRD and PRD for both low and high AOD conditions after the timestep. This may result from leastss humid and morest unstable atmospheric environments over the BTH than over the other two among the three urban clusters (as shown in Section 4.2).

Over the ECS, in both low and high AOD conditions, CDR, CF and CTP decreases during the timestep while COT and CWP increase (see Figure 45). For high AOD conditions, the variations of the cloud properties (CDR, COT and CWP) during the timestep are similar to those for low AOD conditions (Figure 5). Furthermore, it appears that COT and CWP increase more at low AOD than at high AOD. Having a closer look at the CF/CTP variation in both low and high conditions over ocean, we can find that CF decreases (CTP increases) in low AOD conditions and CF increases (CTP decreases) in high AOD conditions over ocean, albeit not over ECS.

In general, the variations over 3 hours in COT and CWP in cloud properties over land are similar to those over ocean for both low and high AOD conditions over 3 hours. Another significant similarity is that CF decreases for low AOD conditions over land and ocean afterduring the 3h timestep. Having a closer look at the CF variation over the YRD and PRD, we can findsee that CF increases in high AOD conditions afterduring the 3h timestep. It infersThis implies that the variation amplitude of CF may depend on the initial AOD conditions. The decrease in afternoon cloud cover over ocean confirms that the largest cover for marine clouds is reached early in the morning as was also concluded by (Meskhidze et al., (2009). Two significant differences are found between land and ocean areas. Meanwhile, oneOnea significant difference is found between land and ocean areas, that is,i.e. –in high AOD conditions CDR increases over land but decreases over ocean after during the 3h timestep under high AOD conditions. , another significant

~~difference is that CF decreases (CTP increases) for low AOD condition but CF increases (CTP decreases) for high AOD condition over ocean after the timestep, whereas CF increases (CTP decreases) for both low and high AOD conditions over land after the timestep. We can conclude that the variation of cloud properties after 3 hours depends little on the initial AOD over land, even though differences exist among the urban clusters. The increase in afternoon cloud fraction over land is consistent with previous studies concluding that continental warm clouds are likely to be well developed (Wang et al., 2014; Kourtidis et al., 2015). The decrease in afternoon cloud cover over ocean confirms that the largest cover for marine clouds is reached early in the morning (Meskhidze et al., 2009).~~ Table 2 summaries the differences in cloud properties between the Aqua and Terra overpasses for high and low AOD conditions over land and ocean during the time period 2008-~~2011~~2017~~, respectively~~.

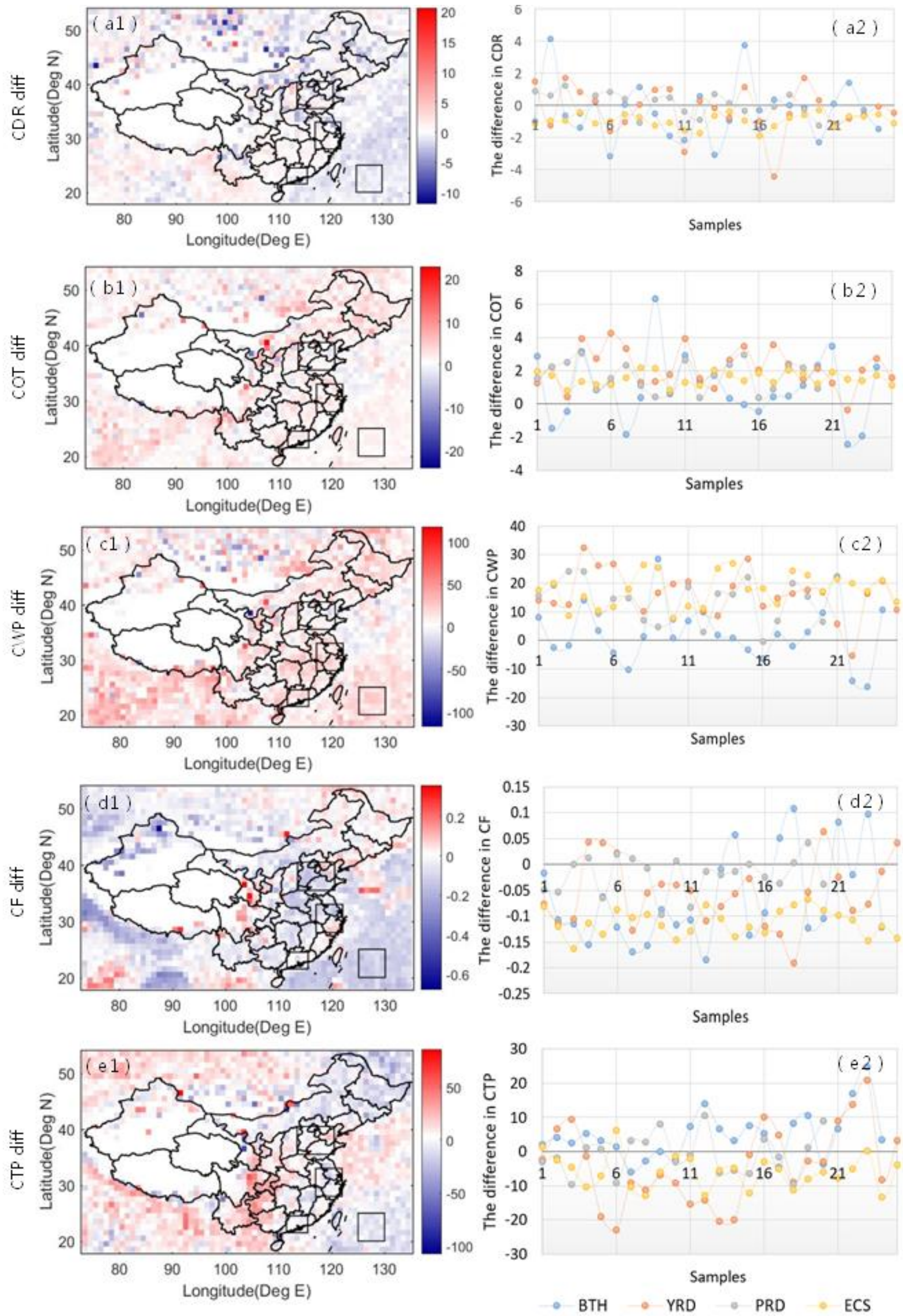


Figure ~~4~~5. Spatial distributions of differences in cloud properties (CDR, COT, CWP, CF and CTP) between Aqua and Terra overpasses (3 hours) for the lowest MODIS/Terra AOD quartiles (left, a1-e1). Sample series of the differences in cloud properties (CDR, COT, CWP, CF and CTP) between the values at the start time and the end time of the timestep for the lowest MODIS AOD quartiles (right, a2-e2).

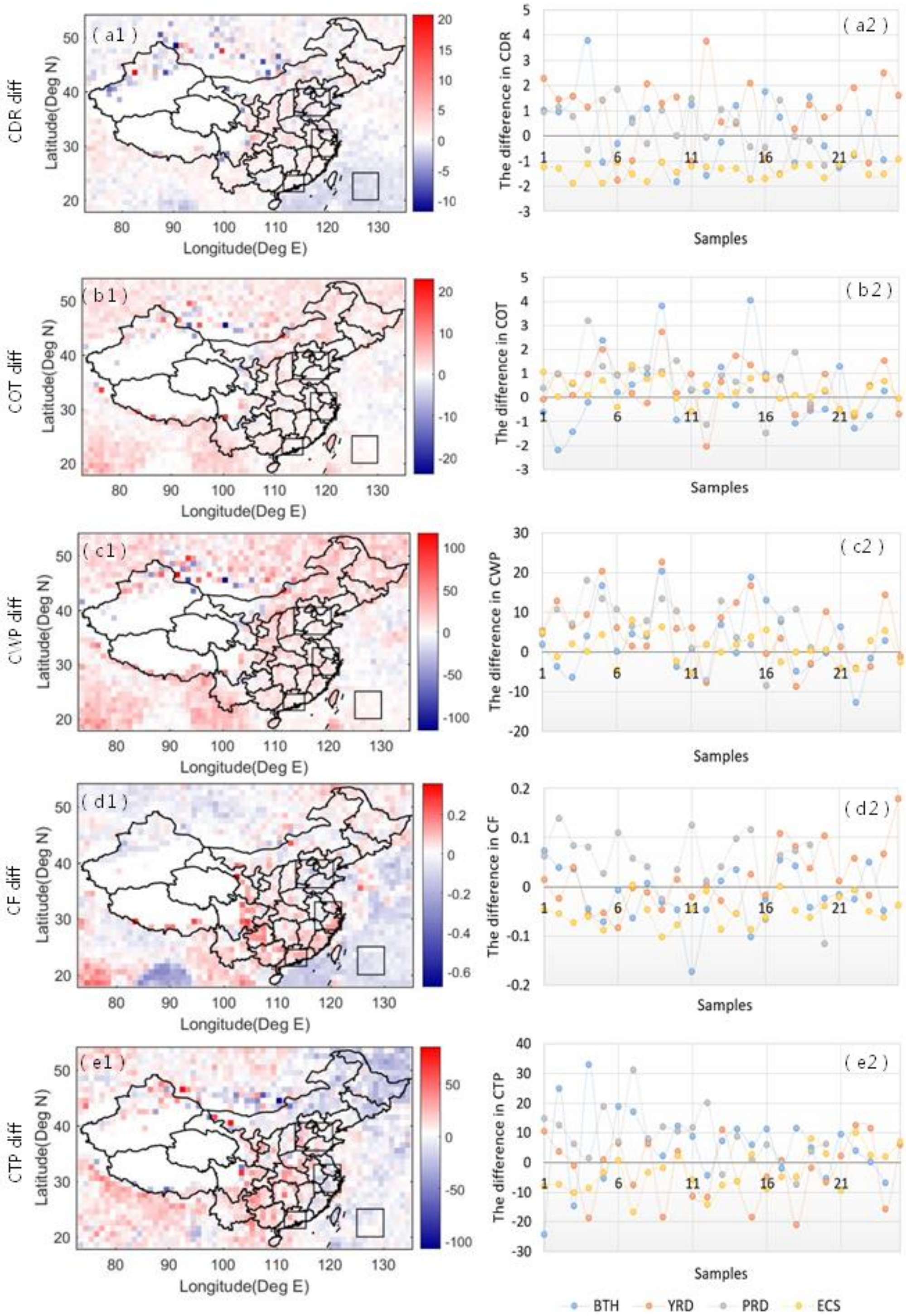


Figure ~~5~~ 6. Spatial distributions of differences in cloud properties (CDR, COT, CWP, CF and CTP) between Aqua and Terra overpasses (3 hours) for the highest MODIS/Terra AOD quartiles (left, a1-e1). Sample series of the differences in cloud properties (CDR, COT, CWP, CF and CTP) between the values at the start time and the end time of the timestep for the highest MODIS AOD quartiles (right, a2-e2).

Table 2 Differences in cloud properties between Aqua and Terra for high and low AOD, over land and ocean.

	Parameters	CDR	COT	CWP	CF	CTP
BTH	L_AOD	-	+**	+	-**	+**
	H_AOD	+	+	+**	-	+**
	d(Cloud_X)	-	-	-	-**	+**
YRD	L_AOD	-	+**	+**	-**	-**
	H_AOD	+**	+**	+**	+	-
	d(Cloud_X)	+	-**	-**	-**	-

PRD	L_AOD	+	+**	+**	-**	-**
	H_AOD	+**	+**	+**	+**	+**
	d(Cloud_X)	+	-**	-**	+**	+**
ECS	L_AOD	-**	+**	+**	-**	-**
	H_AOD	-**	+**	+	-**	-**
	d(Cloud_X)	+**	-**	-**	-**	-

Note: '+' indicates increasing, '-' indicates decreasing and ** at the top right corner of symbol "+" (or "-") denotes that the difference between a change in cloud property and zero is significant (at 95% confidence level).

	~~Land~~			~~Ocean~~		
	~~L_AOD~~	~~H_AOD~~	~~d(Cloud_X)~~	~~L_AOD~~	~~H_AOD~~	~~d(Cloud_X)~~
~~CDR~~	~~+~~	~~+~~	~~--~~	~~-~~	~~-~~	~~--~~
~~COT~~	~~+~~	~~+~~	~~-~~	~~+~~	~~+~~	~~-~~
~~CWP~~	~~+~~	~~+~~	~~-~~	~~+~~	~~+~~	~~-~~
~~CF~~	~~+~~	~~+~~	~~+~~	~~-~~	~~+~~	~~~~~
~~CTP~~	~~-~~	~~-~~	~~-~~	~~+~~	~~-~~	~~--~~

~~注：'+' indicates increasing, '-' indicates decreasing, '~' indicates the response of cloud properties to the increasing AOD is not significant or vague. '--' indicates the difference between the mean change in cloud properties (CF, COT, CWP, CDR and CTP) of the low and high AOD conditions over the timestep is small. L_AOD and H_AOD represent the low and high aerosol conditions, respectively.~~

~~4.3 The difference between the mean changes in cloud properties for low and high AOD over the timestep~~

The differences between the mean changes in cloud properties (CF, COT, CWP, CDR and CTP) between the Terra and Aqua overpasses in high and in low AOD conditions (d (Cloud_X) as defined in ~~section~~ Section 3.2) are investigated to identify the effect of aerosol particles on the cloud properties. Figure ~~6~~ 7 shows the differences between the mean change in cloud properties at low and high AOD conditions during the two observations at 10:30 and 13:30.

Figure ~~6~~ 7 shows that the values of d(CDR) ~~vary around zero~~ over the three urban clusters are not mostly positive or negative, which indicates that ~~during~~ in high ~~and low~~ AOD conditions over land the ~~change~~ variation in CDR during the three hours between the MODIS/Terra and Aqua overpasses is similar. Over the ECS the values of d(CDR) ~~also vary around zero~~~~is~~are ~~negative~~positive, which indicates that the CDR ~~of~~ in high AOD conditions decreases ~~as~~ much more than during low AOD conditions~~as that of low AOD~~ over ocean. Wang et al. (2014) also reported ~~that~~a negative correlation between CDR ~~is negatively associated with~~and AOD over the ECS, ~~agreeing~~in accordance with the Twomey effect. Furthermore, CDR tends to be smallest in polluted and strong-inversion environments, an outcome in good agreement with the findings of Matsui et al. (2006). Most of the d(COT) values are negative over the four regions, especially for the YRD, PRD and ECS. This shows that the COT increases less in high AOD conditions than in low AOD conditions, over both land and ocean, which is contrast with the findings of Meskhidze et al. (2009). Likewise, the values of d(CWP) are almost all negative over the four regions although over the BTH urban cluster the values ~~are less~~ ~~is~~are not clear~~negative than over the other clusters and a number of positive values is observed~~. This indicates that in high AOD conditions the CWP increases less during the timestep than in~~at~~ low AOD conditions, a result in accordance with the conclusion that higher LTS is linked with a slightly lower CWP (Matsui et al., 2006). We can conclude that the variation trend of COT and CWP after 3 hours depends little on the initial AOD, but the initial AOD conditions can affect the amplitude of variation of COT and CWP. Meanwhile, the values of d(CF) are ~~larger~~ smaller than zero over the ~~BTH urban cluster and the~~ ECS. This shows that the cloud fraction in high AOD conditions over the ~~BTH increases and~~ ECS decreases ~~much more~~less than that in low AOD conditions. However, Meskhidze et al. (2009) found that an increase~~d~~ of the ~~–~~ aerosol concentration may lead to enhanced reduction of afternoon cloud coverage and optical thickness for marine stratocumulus regions off the coast of California, Peru, and southern Africa. Therefore, the connection between AOD and variation of cloud cover could be a response to regional-scale changes in aerosol covarying with meteorological conditions. The value of d(CF) is overall positive over ~~land~~the ~~YRD and~~ PRD, which indicates that over the ~~YRD and~~ PRD~~land~~ in high AOD conditions the cloud cover increases much more than the cloud cover decreases in low AOD conditions. ~~However, compared with the variation of d(CF) over the BTH, the variations of d(CF) over the YRD, PRD and ECS regions show a less clear pattern with different behaviors.~~ Mauger and Norris (2007) have shown that scenes with large AOD and large cloud fraction experienced greater LTS. ~~–~~As regards CTP, we find that the values of d(CTP) are positive over the BTH and PRD urban cluster, but the values of d(CTP) over the other two regions do not show a clear pattern ~~negative over the YRD and ECS regions~~. ~~It~~This indicates that in high AOD conditions over the PRD region the CTP increases much more than the CTP decreases in low AOD conditions. ~~In addition, the behavior d(CTP) over the BTH urban cluster is variable with both negative and positive values. Overall, even though there are large variations of d(CTP) with increasing AOD over the three urban clusters, it seems that the value of d(CTP) is negative over land, indicating that in high AOD conditions over land the CTP decreases less than in low AOD conditions.~~ We can conclude that the variation in d(Cloud_X) is different for continental and oceanic clouds. This applies to CDR, cloud fraction (CF) and CTP, but not to COT and CWP.

Table 2 summarizes the differences between the mean changes in cloud properties for low and high AOD over the timestep of 3 hours.

Based on the above findings, we conclude that over the ECS the values of CDR, CWP and CTP are smaller but the values of COT and CF are larger in high AOD conditions. After the 3 hours timestep, CDR, CF and CTP become smaller, irrespective of the AOD. Furthermore, CDR decreases much more in high AOD conditions but CF and CTP decrease~~s~~ much more in high AOD conditions. In contrast, COT and CWP become larger in both ~~two~~ AOD conditions, more significantly in low AOD conditions. Over the urban clusters, COT and CWP also increase over the timestep in both AOD conditions, especially for the low AOD condition. For CF the values in low AOD conditions decrease ~~in low AOD conditions~~ over the timestep. The CTP change behaves different~~ly~~ among the three urban clusters during the 3 hours.

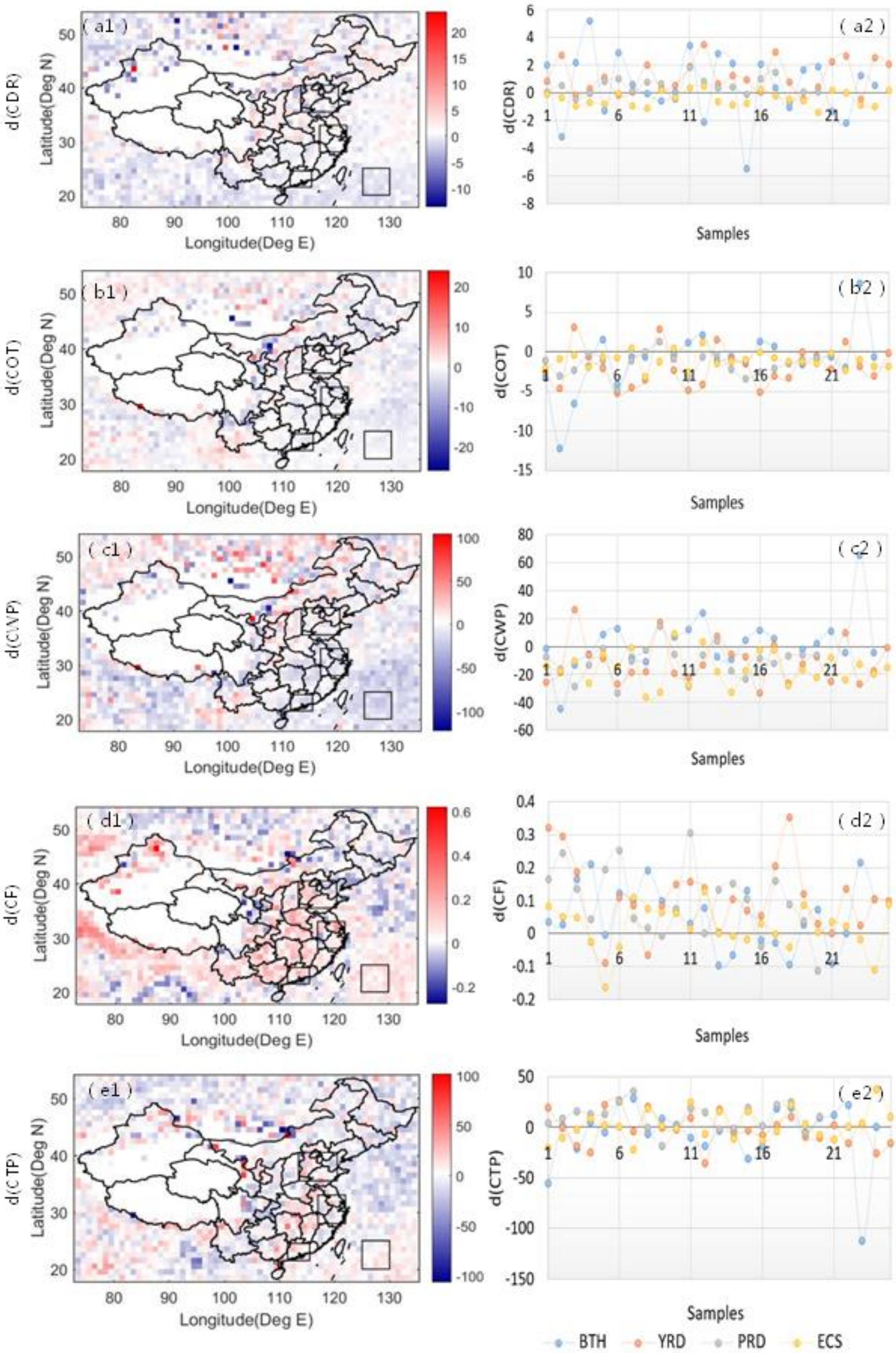


Figure ~~6~~ 7 Spatial distributions (left, a1-e1) and sample time series (right, a2-e2) of d(Cloud_X) (as defined in sect. 3.2) for CDR, COT, CWP, CF and CTP over Eastern China during the time period 2008-~~2011~~2017.

4.4 Meteorological effects

In order to explore the initial meteorological effects on the correlations between AOD and the cloud fraction, we determine the difference in mean cloud parameters between the high and low AOD conditions at the end of the timestep (d(Cloud_X)) in meteorological variable space rather than in longitude-latitude space. Therefore, we define high and low AOD as the highest and lowest quartile for each bin of the meteorological parameters, respectively. Figure ~~7~~ 8 shows the effect of meteorological factors (PVV, RH, ~~LST~~ LTS and initial cloud fraction) on the d(CF) when the cloud cover increases (ΔCloud_X>0) under both low and high AOD conditions over land after the 3 hours timestep. Figure 9 shows the effect of meteorological factors on the d(CF) when the cloud cover deceases (ΔCloud_X<0) under both low and high AOD conditions over land after the 3 hours timestep ~~over land~~. From both figures we find that almost all ~~the values of~~ d(CF) values are positive, indicating that the variations of CF are larger in high AOD than that in low AOD after the 3 hours timestep~~. over land~~.

The PVV, a measure of dynamic convection strength, is very important for cloud formation. The presence of upward motion, as indicated by negative PVV, can enhance the interaction between aerosol particles and clouds as it promotes vertical mixing of the aerosol particles and thus reach the cloud condensation level where they grow into cloud droplets~~makes the ambient environment favorable for cloud formation, and vice versa~~ (Jones et al., 2009). Figure ~~7~~8(a) shows that the d(CF) ~~increases~~ decreases with the PVV over the range from -0.05 Pa s^{-1} to 0.05 Pa s^{-1} ~~when the PVV is negative~~as cloud cover increases in both ~~two~~ conditions over the 3 hours timestep. ~~In contrast, the d(CF) decreases with the PVV when the PVV is larger than zero.~~ This indicates that the weaker downward motion and stronger upward motion of air parcels makes the difference between the increment of cloud cover in high ~~AOD conditions~~ and ~~in~~ low AOD conditions larger: I~~, i~~n other words, the increase rate of cloud cover is larger for high AOD under stronger upward motion of air parcels. Jones et al. (2009) ~~revealed~~showed that stronger upward motion of air parcels can promote the cloud formation in both high and low AOD conditions, but they did not report the increase rate of cloud formation in both ~~two~~ AOD conditions. ~~can enhance the cloud cover much more in conditions with high AOD than in conditions with low AOD.~~ While cloud cover decreases in both ~~two~~ conditions over the 3 hours timestep, Figure 9(a) shows that the d(CF) increases with the PVV over the range from -0.05 Pa s^{-1} to 0 Pa s^{-1} and decreases with the PVV over the range from 0 Pa s^{-1} to 0.05 Pa s^{-1}. This indicates that the decrease rate of cloud cover is smaller for high AOD both under stronger upward motion of air parcels and stronger downward motion of air parcels. Outside~~ of~~ thise range of PVV values the relationship becomes harder to determine due to the reduc~~tion in~~ed data volume~~s~~ in both cases. ~~However, the relative increase in cloud cover is smaller in the presence of downward motion of air parcels in high AOD environment than in low AOD environment.~~ Figure ~~7~~8(b) shows that the d(CF) decreases with increasing RH when RH is lower than ~~92~~20%. This implies that the increase rate of cloud cover is smaller for high AOD ~~cloud cover increases much more in low AOD environment than in high AOD environment~~ with increasing RH. However, when RH is larger than 20~~92~~%, the increase rate of cloud cover is larger for high AOD with increasing RH. A~~a~~n ~~strong~~ increase of d(CF) occurs due to activation of CCN and formation of clouds (Feingold et al., 2003; Liu et al., 2017). It should be noted that the variation of d(CF) with increasing RH above around 80% is uncertain as the sample sizes of high and low AOD conditions are small. ~~On the contrary~~In contrast, the ~~values of~~ d(CF) values become~~s~~ smaller with increasing RH over the whole RH range (See Figure 9(b)), indicating that the decrease rate of cloud cover is smaller for high AOD than that for low AOD with increasing RH.

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). ~~A positive LTS is associated with a stable atmosphere in which vertical mixing is prohibited; negative PVV indicates local upward motion of air parcels.~~ Low LTS represents an unstable atmosphere and high LTS represents a stable atmosphere. Both Figure ~~7~~8(c) and Figure 9(c) show~~s~~ that the d(CF) increases and then decreases with increasing LTS when LTS is lower than 20~~27~~, but increases ~~decreases~~ with increasing LTS for higher values (LTS >20). ~~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 favors the development of a low cloud cover, especially in an environment with high AOD concentrations.~~ However, the sample sizes of high and low AOD conditions are ~~small~~extremely disproportionate when LTS is ~~higher~~ larger than~~ 27~~ 20. Therefore, it is difficult to reach a conclusion from the relationship between d(CF) and LTS ~~is uncertainhard to make a conclusive determination~~ when LTS is larger than 20. Figure ~~7~~8(d) shows ~~that there is weak~~ a strong negative relationship between d(CF) and initial cloud fraction. ~~The effect of initial cloud fraction on d(CF) is not clear.~~The d(CF) increases with increasing initial cloud cover, even though the data volume becomes smaller over the range from 0 to 1.0. ~~It infers~~This implies that the increase rate of cloud cover becomes smaller for high AOD with an increas~~inge~~ of the initial cloud cover~~inconditions~~. Likewise, Figure 9(d) also shows that d(CF) decreases with increasing initial cloud cover, indicating that the decrease rate of cloud cover becomes larger for high AOD with increasing initial cloud cover. This phenomenon is different from the observ~~ation of~~ed weak relationship between d(CF) and initial cloud fraction ~~of~~in the oceanic shallow cumulus regime (Gryspeerdt et al., 2014). It may ~~be~~ result from the combination of above two cases.

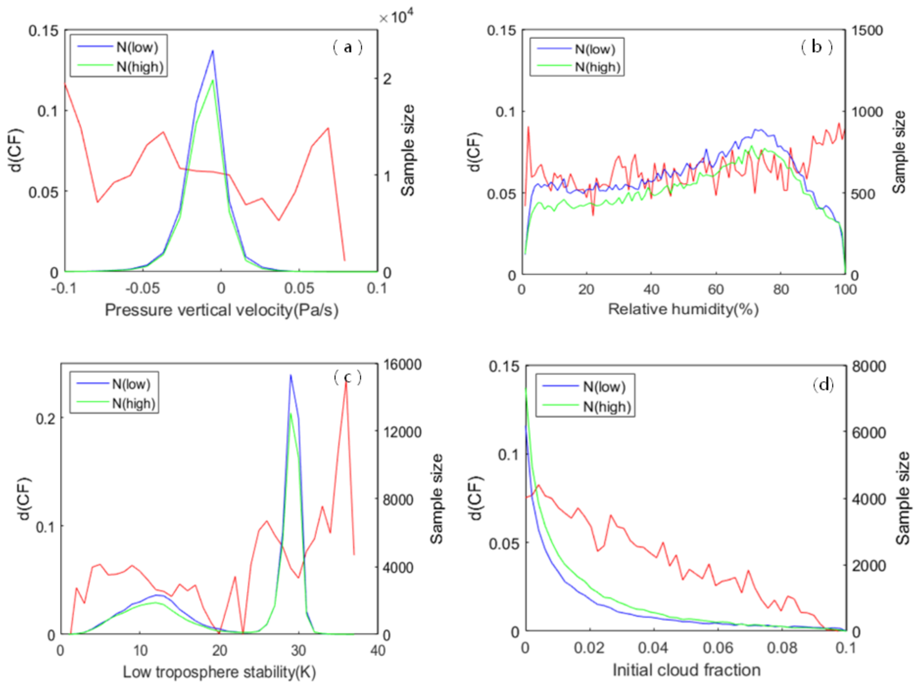


Figure ~~7~~ 8. Variation of d(CF) (red) as function of initial meteorological parameters and cloud fraction for warm clouds when the cloud cover increases under both low and high AOD conditions over land after the 3 hours timestep ~~over land~~. The distribution of points for low (blue) and high (green) AOD as a function of meteorological parameters is shown by the solid lines. This plot is composed from MODIS data (including Terra and Aqua) for all warm cloud points over the years 2008-~~2011~~2017. Meteorological parameters are plotted along the horizontal axis, the left vertical axis denotes d(CF) and the right vertical axis denotes the number of high and low AOD samples.

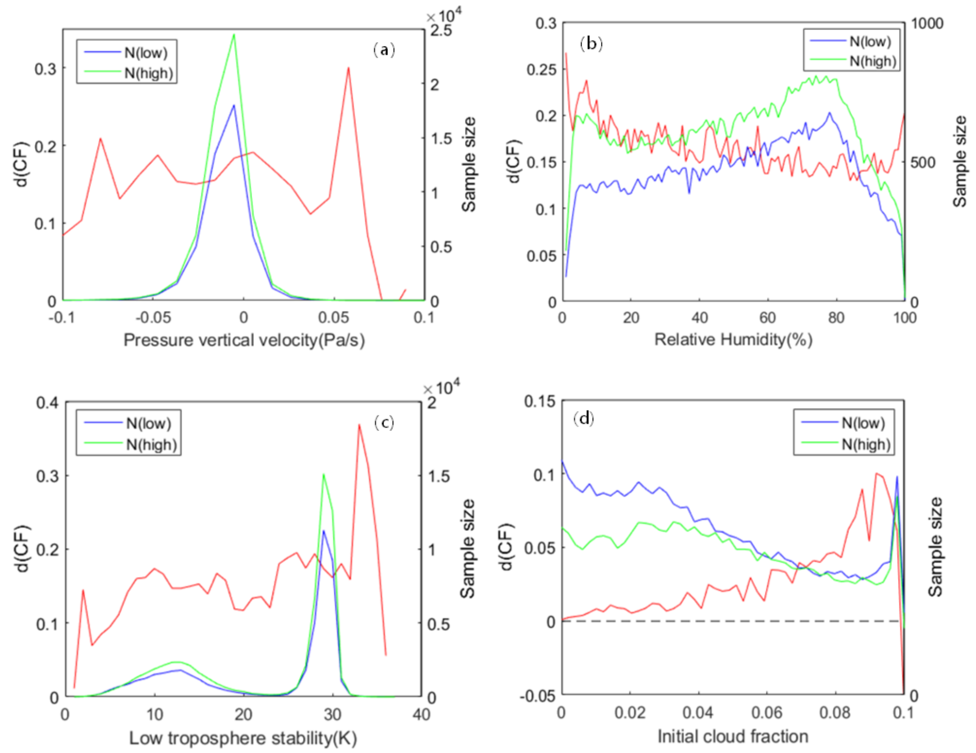


Figure 9 The same as Fig. 8 but for warm clouds when the cloud cover decreases under both low and high AOD conditions over land after the 3 hours timestep ~~over land~~.

5 **Conclusions**

The large 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). In this work, based on the near-simultaneous aerosol and cloud retrievals provided by MODIS, together with the ERA Interim Reanalysis data, we investigated the effect of aerosol loading, using ~~as indicated by~~ AOD as a proxy, on aerosol-cloud interactions. Aerosol-cloud interaction was studied over three major urban clusters in eastern China and over one area over the Eastern China Sea. These four areas are representative of different climatic regions and pollution levels. Data over these four study areas were collected for the years 2008 to ~~2011~~2017, and analyzed in a statistical sense ~~using statistical methods~~. Both MODIS/Terra and MODIS/Aqua data were used to study the difference ~~of~~ in cloud properties between the morning and the early afternoon, i.e. with a time difference of 3 hours.

In order to reduce ~~the~~ differences ~~of~~ in the initial ~~conditions~~ distributions of cloud and meteorological parameters between high and low AOD conditions at the start of the timestep, normalized histograms of these ~~cloud properties and meteorological~~ parameters were made for high and low AOD conditions following the method described by Gryspeerdt et al., (2014). After that, the difference between cloud properties (CDR, COT, CWP, CF and CTP) in high and low AOD conditions during the Terra overpass at 10:30 LT for each 1°×1° grid was investigated. We looked at statistics for the 10~~4~~-years dataset and found that different regions with various aerosol emission levels, aerosol types and different climate characteristics show different patterns of ACI. The ACI is more complex over land (BTH, YRD and PRD) than over ocean (ECS). Next, the mean change in cloud properties during the 3 hours between the observations in low and high AOD conditions, as provided by the differences in the observations by MODIS/Terra (morning) and MODIS/Aqua (afternoon) overpasses, were examined and differences were analyzed. The results show that the COT and CWP over land and ocean were increased after the 3 hours timestep ~~over land and ocean~~, irrespective of the initial AOD conditions. ~~in low and high AOD conditions the variation of cloud properties between the two observations behave similarly (increase or decrease). In general, the variation of cloud properties over the urban clusters is similar to that over ocean. Two significant differences are found between land and ocean areas. One is that CDR increases over land but decrease over ocean after the 3-hour period, another significant difference is that CF decreases (CTP increases) for low AOD condition but CF increases (CTP decreases) for high AOD condition over ocean during the timestep, whereas CF increases (CTP decreases) during that period for both low and high AOD conditions over land.~~ Furthermore, we investigated the difference between the mean change in cloud properties (CDR, COT, CWP, CF and CTP) in low and high AOD conditions between the two observations. We found that the variation in d(Cloud_X) is different for continental and oceanic clouds. This applies to CDR, cloud fraction and CTP, but not to COT and CWP. Both COT and CWP increase over land and ocean after the timestep, irrespective of the AOD. The variation trend of COT and CWP after 3 hours depends little on the initial AOD, but the initial AOD conditions can affect the amplitude of variation of COT and CWP.

Constrained by relative humidity and boundary thermodynamic and dynamic conditions, the variation of d(CF) in response to aerosol abundance over land was also analyzed. ~~There are~~ T~~t~~wo cases ~~a~~were considered: (1) when the cloud cover increases under both low and high AOD conditions after the 3 hours timestep; (2) when the cloud cover decreases under both low and high AOD conditions after the 3 hours timestep. From both ~~two~~ cases, we find that almost all ~~the values of~~ d(CF) values are positive, indicating that the variations of CF are larger in high AOD than that in low AOD after the 3 hours timestep. The results show that cloud cover increases much for high AOD under stronger upward motion of air parcels;

Meanwhile, the increase rate of cloud cover is larger for high AOD with increasing RH when RH greater than 20%. ~~scenes with large cloud fraction experience large AOD and stronger upward motion of air parcels and large RH. of~~ With regarded to the effect of LTS on the change of cloud cover, scenes with large cloud fraction ~~change~~variation experience large AOD and large LTS when LTS smaller than 10. Conversely, scenes with smaller cloud fraction ~~change~~variation experience large AOD and large LTS when LTS larger than 10 and smaller than 20. We also find that smaller increase rate of cloud fraction occurs when scenes experience larger AOD and larger initial cloud cover~~ins~~.~~the presence of upward motion of air parcels can enhance the cloud cover much more in high than in low AOD conditions. In contrast, the cloud cover increases much more with increasing RH in clean atmospheric conditions than in polluted atmospheric conditions. Meanwhile, stable atmospheric conditions favor the development of a low cloud cover, especially in high AOD conditions. A statistical analysis of the relation between d(CF) and initial cloud fraction shows a weak negative relationship between d(CF) and initial cloud fraction.~~

In summary, whilst we have reduced the error due to meteorological effects on aerosol retrieval, meteorological covariation with the cloud and aerosol properties is harder to remove. As aerosol-cloud interaction is a complex problem, it is important to synergistically use multiple observation products and atmospheric models to explore the mechanisms of aerosol-cloud interaction. Therefore, further analysis can be carried out in future work.

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Reply to comments on "Satellite-based estimate of the variability of warm cloud properties associated with aerosol and meteorological conditions"

October 30, 2018

We thank the reviewer's thoughtful comments which are helpful not only for this manuscript but also for our future research. Our replies for all the comments are shown below.

Major points

1. Comments: (1) The authors make use of a technique previously used to investigate possible links between aerosol and cloud fraction, extending it to look at the development of other cloud properties. A key part of this method involves making sure that the starting state similar as possible for high and low aerosol environments and then investigating the difference between them. If this method works as intended, the mean change in cloud properties over the timestep should be a function only of local meteorology and there should be no difference in the cloud properties between the high and low aerosol populations at the start time. I am therefore unclear what is being shown in section 4.1, where a difference apparently exists. Are the authors following the method of Gryspeerdt et al (2014), or have they created a new method? If the authors are just looking at the relationship between AOD and cloud properties, how have they accounted for the impact of local meteorology (e.g. Quaas et al, ACP, 2010)?

Answer: Normalised histograms of cloud properties for the high and low AOD populations are made for the whole region (Section 3.1), because the data volume based on each 1° x 1° location is relatively small. However, the difference between the cloud properties for low and high AOD at the start time is based on each 1° x 1° location (Section 4.1). So the difference of the cloud properties between the low and high AOD at the start time still exist and is not zero. In order to make the reader understand, text was added as follows.

Page 5 lines 37-39 and page 6 lines 1-2, : Text was added as:' Note that here and in the following sections, normalised histograms of cloud properties for the high and low AOD populations are made for the whole region (Section 3.1), because the data volume based on each 1° x 1° location is relatively small. However, the difference between the cloud properties for low and high AOD at the start time is based on each 1° x 1° location (Section 4.1). So the difference of the cloud properties between the low and high AOD at the start time is not zero.'

Page 9, line 15-17: Text was added as:' Although normalized histograms of meteorological parameters are made for high and low AOD conditions at the start time, the normalization described in Sect. 3.1 is based on the whole region. Differences in meteorological conditions may still occur between each 1° x 1° grid cell.'

Meanwhile, in order to consider the effect of meteorological conditions on the relationship between aerosol and cloud further, we analyze the meteorology of the different regions in Section 4.2 (see page 9-10). This new Section 4.2 "The meteorology of the four target regions" reads:

4.2 The meteorology of the four target regions

The meteorological and aerosol effects on clouds are reported to be tightly connected, and this connection must be accounted for in any study of aerosol-cloud interactions (Stevens and Feingold, 2009; Koren et al., 2010). Although normalized histograms of meteorological parameters are made for high and low AOD conditions at the start time, the normalization described in Sect. 3.1 is based on the whole region. Differences in meteorological conditions may still occur between each 1° x 1° grid cell. In this study, we analyze the meteorology of the different regions, in support of the interpretation of the regional variation of the relationships between aerosols and clouds.

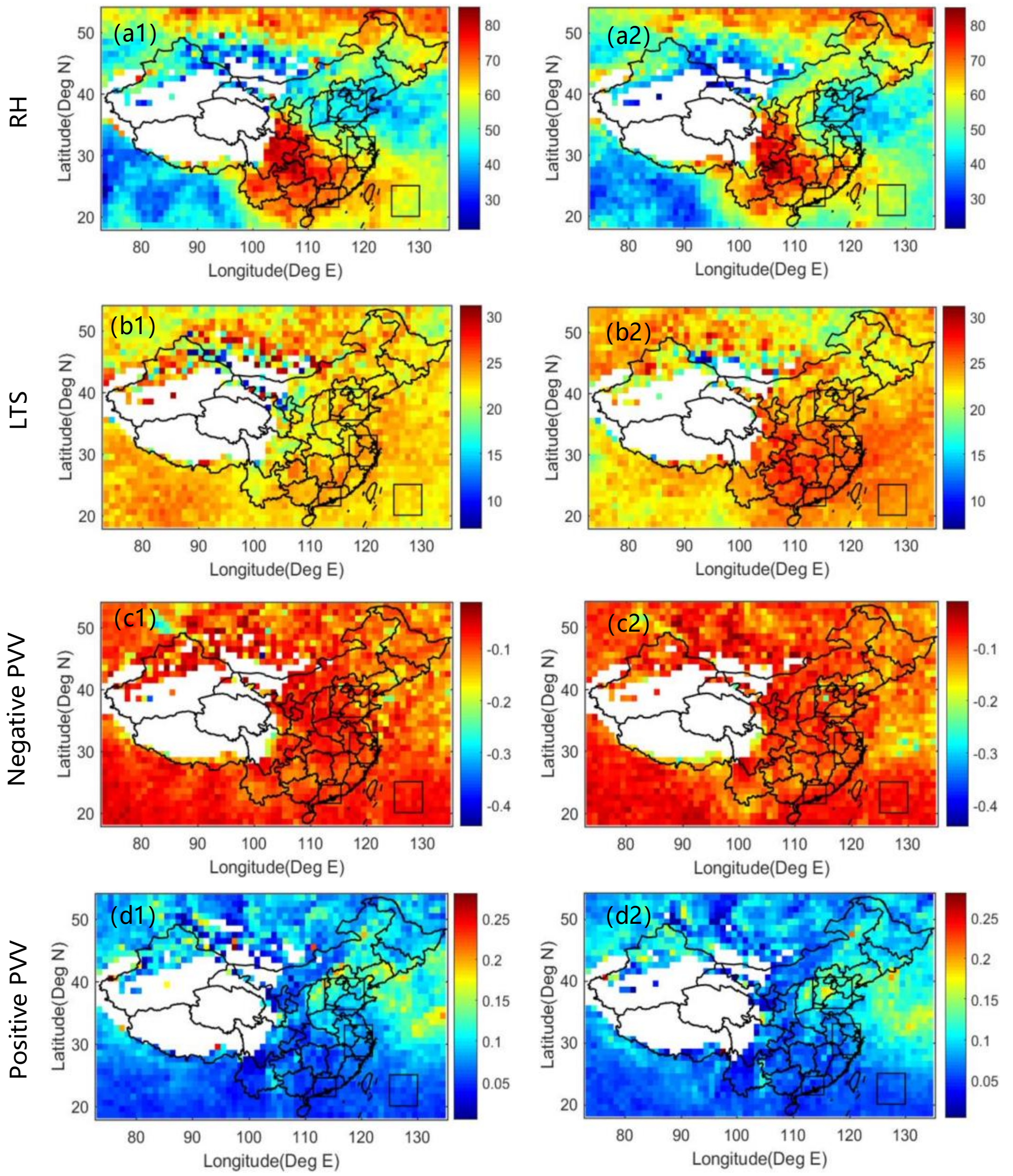

Figure 4 Spatial distributions of meteorological parameters (top to bottom: RH, LTS, positive PVV and negative PVV) at the start time of the timestep (MODIS/Terra) for low AOD conditions (left, a1-d1) and for high AOD conditions (right, a2-d2). All the data are averaged over all years between 2008 and 2017.

The spatial variations of the aerosol and cloud properties over the four regions, averaged over the years 2008-2017, are shown in Fig. 4. Over the urban clusters, we can see an increasing north–south pattern in RH and LTS, with the lowest values found in the PRD. For the negative PVV, the spatial distributions for the low and high AOD situations are remarkably similar, with the highest values over the BTH and decreasing toward the south to near zero over the PRD. In contrast, the positive PVV is smallest over the BTH, with little variation over the study area. Overall, the meteorological parameters over the YRD and PRD are similar to those over the ECS, irrespective of the AOD. Furthermore, the LTS is significant larger in the high AOD conditions for all the four regions. Zhao et al. (2006) proposed that the enhancement in the atmospheric stability tends to depress upward motion and precipitation, leading to an increase in aerosol particles. The spatial distributions of both positive and negative PVV in the low AOD conditions are similar to those in the high AOD conditions.

2. Comments: (2) Similarly, it is not clear what section 4.2 is showing. While the title states that it is discussing the 'mean change', it is apparently also investigating the difference between high and low AOD. If this is the case, could it not be merged with section 4.3, which is explicitly about the difference in relation to the aerosol environment? I would expect that the difference in the development between the regions would be a function of local meteorology. If 4.2 is intended to be about the mean cloud development, perhaps it could be used to better describe the meteorology of the different regions, helping the interpretation of the regional variation of the results in section 4.3.

Answer: Yes, we agree with your suggestions. Section 4.2 was merged with Section 4.3 (as new Section 4.3, see pages 10-15), explicitly examining the difference of cloud properties in relation to aerosol environment. Furthermore, new Section 4.2 was added (see response to question 1) to describe the meteorology of the four target regions, in support of the interpretation of the regional variation of relationship between aerosol and cloud (see page 9-10 in the revised manuscript).

3. Comments: (3) While this work has the potential for producing interesting results if the method is properly clarified, the results that are currently within the paper are not set in the context of existing work, which makes them difficult to interpret. The results in section 4.3 and not compared to section 4.1 or previous work, meaning that potentially interesting results are missed. As some examples, P13L14 suggests that there is little change in the CDR development as a function of aerosol - this inability to detect the Twomey effect might mean that this method is not suitable for investigating aerosol cloud interactions, or it could mean that changes in CDR proceed via different pathways and timescales than the CF changes observed in Gryspeerdt et al. (2014). Although the difference in results over land and ocean was one of the key results of Gryspeerdt et al (2014), other result are different - this work finds exactly the opposite dCF response to relative humidity (section 4.4). This would again be an interesting result for discussion that is missed as it is not set in context.

Answer: The variation of cloud properties to the aerosol environment has become more clear by reanalyzing all the MODIS C6.1 data for the whole acquisition period between 2008 and 2017, rather than MODIS C5.1 data from 2008 to 2011. This change is shown throughout the revised manuscript (all the figures were changed/modified in this respect). Following the reviewer's comments, the results in Section 4.3 have been linked to Section 4.1 and compared to previous work.

Part of text in Section 4.3 was shown in follows (see page 14-15 in the revised manuscript):

"Figure 7 shows that the values of d(CDR) over the three urban clusters are not mostly positive or negative, which indicates that in high AOD conditions over land the variation in CDR during the three hours between the MODIS/Terra and Aqua overpasses is similar. Over the ECS the values of d(CDR) is positive, which indicates that the CDR in high AOD conditions decreases much more than during low AOD conditions over ocean. Wang et al. (2014) also reported a negative correlation between CDR and AOD over the ECS, in accordance with the Twomey effect. Furthermore, CDR tends to be smallest in polluted and strong-inversion environments, an outcome in good agreement with the findings of Matsui et al. (2006). Most of the d(COT) values are negative over the four regions, especially for the YRD, PRD and ECS. This shows that the COT increases less in high AOD conditions than in low AOD conditions, over both land and ocean, which is contrast with the findings of Meskhidze et al. (2009). Likewise, the values of d(CWP) are almost all negative over the four regions although over the BTH urban cluster the values are not clear. This indicates that in high AOD conditions the CWP increases less during the timestep than in low AOD conditions, a result in accordance with the conclusion that higher LTS is linked with a slightly lower CWP (Matsui et al., 2006). We can conclude that the variation trend of COT and CWP after 3 hours depends little on the initial AOD, but the initial AOD conditions can affect the amplitude of variation of COT and CWP. Meanwhile, the values of d(CF) are smaller than zero over the ECS. This shows that the cloud fraction in high AOD conditions over ECS decreases less than that in low AOD conditions. However, Meskhidze et al. (2009) found that an increase of the aerosol concentration may lead to enhanced reduction of afternoon cloud coverage and optical thickness for marine stratocumulus regions off the coast of California, Peru, and southern Africa. Therefore, the connection between AOD and variation of cloud cover could be a response to regional-scale changes in aerosol covarying with meteorological conditions. The value of d(CF) is overall positive over the PRD, which indicates that over the PRD in high AOD conditions the cloud cover increases much more than the cloud cover decreases in low AOD conditions. Mauger and Norris (2007) have shown that scenes with large AOD and large cloud fraction experienced greater LTS. As regards CTP, we find that the values of d(CTP) are positive over the BTH and PRD urban cluster, but the values of d(CTP) over the other two regions are not significant. It indicates that in high AOD conditions over the PRD region the CTP increases much more than the CTP decreases in low AOD conditions. We can conclude that the variation in d(Cloud_X) is different for continental and oceanic clouds. This applies to CDR, cloud fraction (CF) and CTP, but not to COT and CWP. Table 2 summarizes the differences between the mean changes in cloud properties for low and high AOD over the timestep of 3 hours.

Based on the above findings, we conclude that over the ECS the values of CDR, CWP and CTP are smaller but the values of COT and CF are larger in high AOD conditions. After the 3 hours timestep, CDR, CF and CTP become smaller, irrespective of the AOD. Furthermore, CDR decreases much more in high AOD conditions but CF and CTP decreases much more in low AOD conditions. In contrast, COT and CWP become larger in both AOD conditions, more significantly in low AOD conditions. Over the urban clusters, COT and CWP also increase over the timestep in both AOD conditions, especially for the low AOD condition. For CF the values in low AOD conditions decrease over the timestep. The CTP change behaves differently among the three urban clusters during the 3 hours.”

The sentence in P13L14 in old version manuscript is “Figure 6 shows that the values of d(CDR) vary around zero over the three urban clusters, which indicates that during high and low AOD over land the change in CDR during the three hours between the MODIS/Terra and Aqua overpasses is similar.” The sentence means that there are changes (increase or decrease) of CDR in both AOD conditions after 3 hours timestep, but the variation quantity is similar. So, it doesn’t indicate that this inability to detect the Twomey effect. As Figures 3(a1-a2) show, over the ECS, CDR is smaller at high AOD than at low AOD, which is consistent with Twomey’s effect. In contrast, over the three urban clusters, CDR is larger at high AOD. This behavior has been observed before for warm clouds in conditions with high AOD (Liu et al., 2017) and may result from the intense competition for the available water vapour and the evaporation of smaller droplets as a consequence of the high aerosol abundance over these regions (Yuan et al., 2008; Wang et al., 2014; Tang et al., 2014; Liu et al., 2017).

The effects of initial cloud fraction and meteorological conditions on the change in CF under low and high AOD conditions after the 3 hours timestep over land are also explored. In our new version manuscript, there are two cases are considered: (1) when the cloud cover increases (ΔCloud_X>0); (2) when the cloud cover decreases (ΔCloud_X<0). The d(CF) (see Section 3.2) response to relative humidity is different for both cases (see Section 4.4 in the revised manuscript). However, the results of Gryspeerdt et al. (2014) are based on the combination of the two cases.

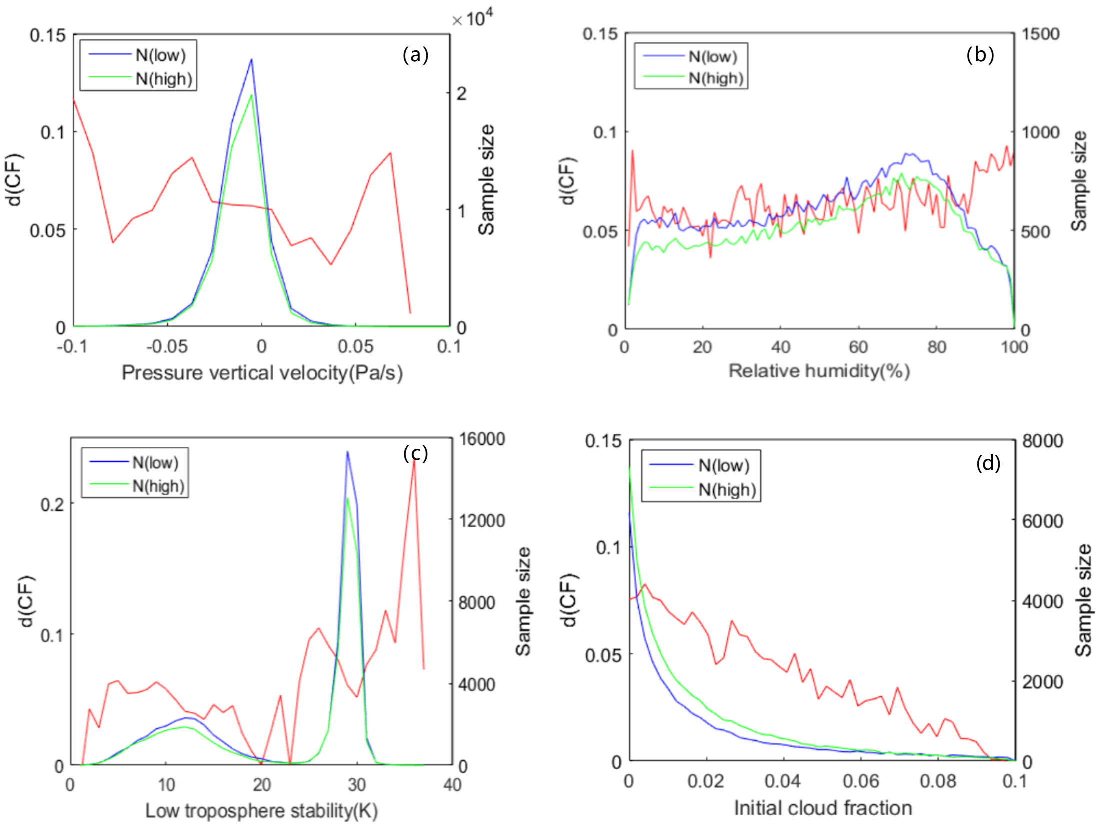

Figure 8. Variation of d(CF) (red) as function of initial meteorological parameters and cloud fraction for warm clouds when the cloud cover increases under both low and high AOD conditions after the 3 hours timestep over land. The distribution of points for low (blue) and high (green) AOD as a function of meteorological parameters is shown by the solid lines. This plot is composed from MODIS data (including Terra and Aqua) for all warm cloud points over the years 2008-2017. Meteorological parameters are plotted along the horizontal axis, the left vertical axis denotes d(CF) and the right vertical axis denotes the number of high and low AOD samples.

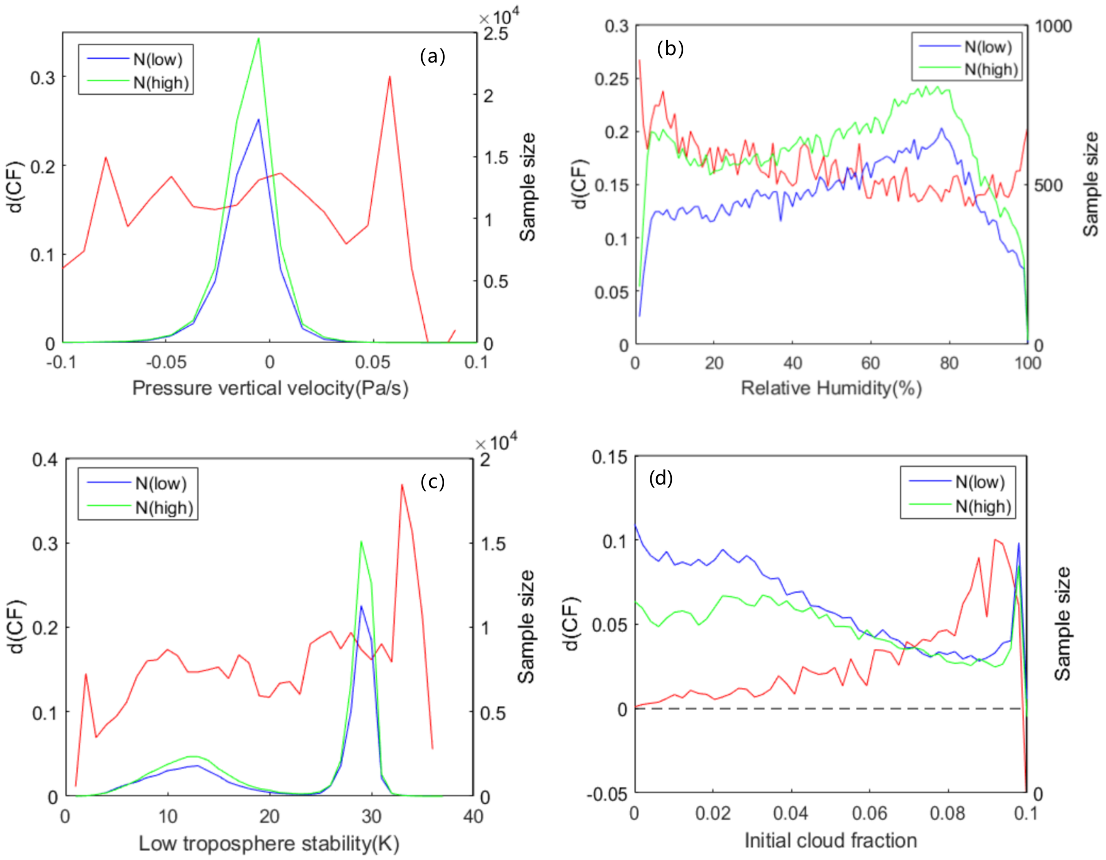

Figure 9 The same as Fig. 8 but for warm clouds when the cloud cover decreases under both low and high AOD conditions after the 3 hours timestep over land.

4. Comments: (4) I am not clear of the purpose of choosing the different regions in this work. They are explained in section 2, but very little reference is made to these meteorological differences later in the paper. Other than noting that the aerosol-cloud relationships are different in these regions, there is little discussion of why there is a difference. As variations have previously been noted in the strength of aerosol-cloud relationships, it would be good to include some discussion as to why they are different. This would help this paper build on the previous literature in this area.

Answer: Yes, following the reviewer's comments, we add the meteorology of the four target regions in new Section 4.2, in support of the interpretation of the regional variation of relationship between aerosol and cloud. Furthermore, we have discussions of those different aerosol-cloud relationships in different regions and gave possible

reasons (see pg.11 lines 12-17 in the revised manuscript).
In order to include some discussion as to why they are different, text was added as: ’From the perspective of considering all urban clusters (BTH, YRD and PRD), both COT and CWP increase over land during the 3 hours timestep for both low and high AOD. Overall, the variation in cloud properties after the timestep over BTH is less significant than over the YRD and PRD for both low and high AOD conditions. This may result from less humid and more unstable atmospheric environments over the BTH than over the other two urban clusters (as shown in Section 4.2).’ in the Page 11, line 12-17.

Specific comments

1. Comments: (1) Page 1, Line 39: Twomey 1974/77?

Answer: We made this change (see page 2, lines 3-4). “Due to interactions with aerosols, the climatic effects of clouds are further complicated (Rosenfeld, 2000; Twomey, 2007)” has been changed to “Due to interactions with aerosols, the climatic effects of clouds are further complicated (Rosenfeld, 2000; Twomey, 1974; Twomey, 1977).”

2. Comments: (2) Page 2, Line 3: a smaller droplet radius does not always result in precipitation suppression, especially if the warm rain frequency is already low (e.g. Muelmenstaedt et al., GRL, 2015)

Answer: “Thus, cloud albedo increases and the smaller cloud droplet effective radius results in the suppression of precipitation, which in turn results in a longer cloud lifetime, and maintaining a larger liquid water path (Albrecht, 1989; Feingold et al., 2001)” has been changed to “Thus, cloud albedo increases and the smaller cloud droplet effective radius in most cases results in the suppression of precipitation, which in turn results in a longer cloud lifetime, and maintaining a larger liquid water path (Albrecht, 1989; Feingold et al., 2001)” in the revised manuscript (see page2, line 8-10).

3. Comments: (3) Page 4, Line 1: Why not use collection 6 data? There is also almost four times as much MODIS daily data available as it being used here. Why has this specific time period been chosen? A larger data record would improve the statistical significance of this work.

Answer: Following the reviewer’s comments, we use collection 6.1 data and reanalyze all the data for the whole acquisition period between 2008 and 2017, rather than C5.1 data from 2008 to 2011. Therefore, the variation of cloud properties to the aerosol environment has been changed and more clear. This issue is shown throughout the revised manuscript (all the figures were changed/modified in this respect).

4. Comments: (4) Page 4, Line 24: Why is aerosol optical depth used? Many previous studies have that it had severe limitations proxy for CCN (e.g. Penner et al, PNAS, 2011)

Answer: The average CCN concentrations show a remarkable correlation to the corresponding AOT values, it provides an easily measured proxy for CCN concentration (Andreae, 2009). Meanwhile, in the present study the use of AI would not be appropriate, because our study is conducted mostly over land areas. This has to do with the use of the Ångström exponent in the derivation of AI, namely, the Ångström exponent is not reliable over land areas. We quote a personal communication with L. Remer (20 June 2010), NASA GSFC: "Ångström over land is not reliable and we recommend strongly not to use it"; hence, AOD is used in our study (Kourtidis et al., 2015).

5. Comments: (5) Page 5, Line 2: 'representative of typical thermodynamic conditions' it is not clear what this means.

Answer: "…which is representative of typical thermodynamic conditions (Klein and Hartmann, 1993)." has been changed to "…which can be regarded as a measure of the strength of the inversion that caps the planetary boundary layer (Klein and Hartmann, 1993; Wood and Bretherton, 2006)" in the revised manuscript (see page 4, line 30-31).

6. Comments: (6) Page 6, Lines 1: Are all parameters considered at the same time? Gryspeerdt et al, also used meteorological parameters normalization.

Answer: Yes, normalized histograms of cloud properties and meteorological parameters are made for high and low AOD conditions following the method described by Gryspeerdt et al. (2014).

7. Comments: (7) Page 6, Line 2: Normalisation by cloud fraction makes the biggest difference in what?

Answer: We made this change (see page 5 lines 34-35). "…even though we find that the normalization for the cloud fraction made the biggest difference by far." has been changed to "Among those cloud properties, this process of normalization has the greatest effect on the cloud fraction and its dependence on aerosol-cloud interaction."

8. Comments: (8) Page 6, Line 2: Does this mean this normalization method is applied throughout this work?

Answer: Yes, the sentence means the normalization method is applied throughout the work. And "In the further analysis, we only take a subset of original data by removing random samples until the histograms are similar." has been changed to "Throughout the work, we only take a subset of original data by removing random samples until the histograms are similar." (see page 5, line 35-36 in the revised manuscript)

9. Comments: (9) Page 6, Line 24: As mentioned earlier should the difference between the cloud properties at the start time not be zero?

Answer: Normalised histograms of cloud properties for the high and low AOD populations are made for the whole region (Section 3.1), because the data volume based on each 1° x 1° location is relatively small. However, the difference between the cloud properties for low and high AOD at the start time is based on each 1° x 1° location (Section 4.1). So the difference of the cloud properties between the low and high AOD at the start time is not zero (see response to question 1 in major points section).

10. Comments: (10) Page 7, Line 7, Perhaps also Yuan et al, ACP, 2008 (Increase of cloud droplet size with aerosol optical depth: An observation and modeling study, 10.1029/2007JD008632)

Answer: We made this change (see page 7, line 6-8). "…may result from the intense competition for the available water vapour and the evaporation of smaller droplets as a consequence of the high aerosol abundance over these regions (Wang et al., 2014; Liu et al., 2017)." has been changed to "…may result from the intense competition for the available water vapour and the evaporation of smaller droplets as a consequence of the high aerosol abundance over these regions (Yuan et al., 2008; Tang et al., 2014; Wang et al., 2014; Liu et al., 2017)."

11. Comments: (11) Page 7, Line 22: Many previous studies have shown links between aerosol and cloud properties over China but it might be good to know why these relationships are different.

Answer: We made this change (see page 9 lines 1-8). Text are added as: "Some links between aerosol and cloud in the four regions are different from those of previous studies over China (Wang et al., 2014; Tang et al., 2014; Kourtidis et al., 2015; Liu et al., 2017), which might be due to the use of different data sets (MODIS C6.1 versus older versions), hypothesis and target areas characterized by complex aerosol compositions and varying meteorological conditions. Overall, the result implies that the interaction between aerosol particles and clouds is more complex and of greater uncertainty over land (BTH, YRD and PRD) than over ocean (ECS). Jin and Shepherd (2008) also noted that aerosol affect clouds more significantly over ocean than over land. They suggested that dynamic processes related to factors like urban land cover may play at least an equally critical role in cloud formation."

12. Comments: (12) Figure3: What is this sample time series?

Answer: Samples are collected from the pixels of the difference in cloud properties that covering the four regions and randomly as shown in the Figure3.

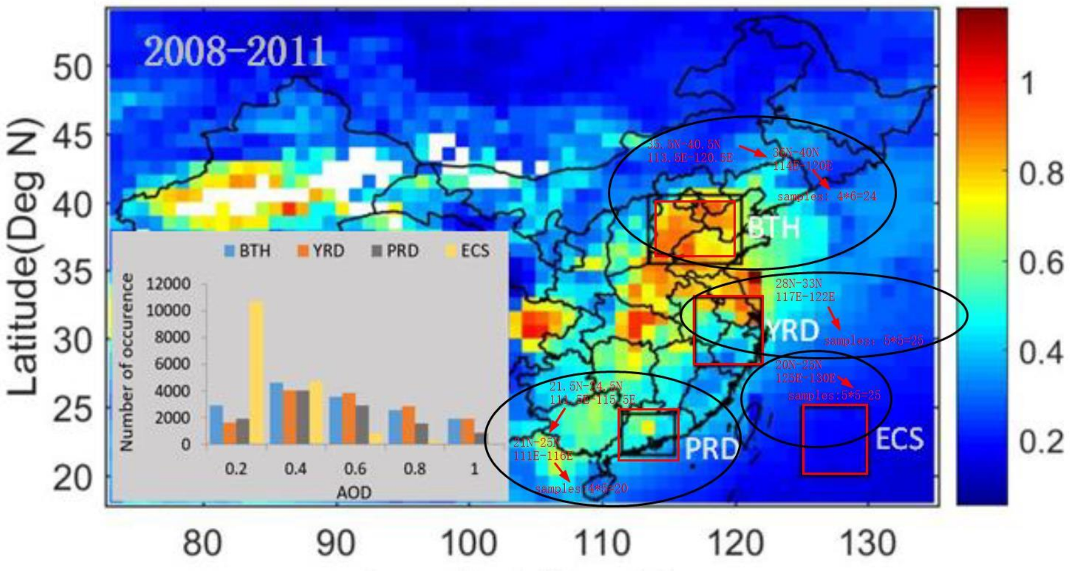

Figure A map of showing samples are collected in the four target regions.

13. Comments: (13) Page 10, Line 4: If the variation of cloud properties depends little on the initial AOD, does that not mean that section 4.3 should show no results? This would be in contrast to previous studies.

Answer: We made this change (see page 11, line 12-18 in the revised manuscript). The sentence "We can conclude that the variation of cloud properties after 3 hours depends little on the initial AOD over land, even though differences exist among the urban clusters" has been removed, which is not a correct conclusion. Further, the variation of cloud properties to the aerosol environment using different data sets (MODIS C6.1 versus older versions), we find that both COT and CWP increase over land during the 3 hours timestep for both low and high AOD. Overall, the variation in cloud properties after the timestep over BTH is less significant than over the YRD and PRD for both low and high AOD conditions. This may result from the less humid and most unstable atmospheric environments over the BTH than over the other two urban clusters (as shown in new Section 4.2). Over the ECS, in both low and high AOD conditions, CDR, CF and CTP decrease during the timestep while COT and CWP increase (see Figure 5 in the revised manuscript).

14. Comments: (14) Page 13: As there have been several previous studies looking at aerosol and cloud relationships, it would be good to set these results in context of previous work.

Answer: We made this change (see page 14 lines 8-37 and page 15 lines 1-9). The variation of cloud properties to the aerosol environment has been more clear by reanalyzing all the MODIS C6.1 data for the whole acquisition period between 2008 and 2017, rather than MODIS C5.1 data from 2008 to 2011. This issue is shown throughout the revised manuscript (all the figures were changed/modified in this respect). Following the reviewer's comments, the results in Section 4.3 have been linked to Section 4.1 and compared to previous work.

Part of text in Section 4.3 was shown in follows (see page 14-15 in the revised manuscript): “Figure 7 shows that the values of d(CDR) over the three urban clusters are not mostly positive or negative, which indicates that in high AOD conditions over land the variation in CDR during the three hours between the MODIS/Terra and Aqua overpasses is similar. Over the ECS the values of d(CDR) is positive, which indicates that the CDR in high AOD conditions decreases much more than during low AOD conditions over ocean. Wang et al. (2014) also reported a negative correlation between CDR and AOD over the ECS, in accordance with the Twomey effect. Furthermore, CDR tends to be smallest in polluted and strong-inversion environments, an outcome in good agreement with the findings of Matsui et al. (2006). Most of the d(COT) values are negative over the four regions, especially for the YRD, PRD and ECS. This shows that the COT increases less in high AOD conditions than in low AOD conditions, over both land and ocean, which is contrast with the findings of Meskhidze et al. (2009). Likewise, the values of d(CWP) are almost all negative over the four regions although over the BTH urban cluster the values are not clear. This indicates that in high AOD conditions the CWP increases less during the timestep than in low AOD conditions, a result in accordance with the conclusion that higher LTS is linked with a slightly lower CWP (Matsui et al., 2006). We can conclude that the variation trend of COT and CWP after 3 hours depends little on the initial AOD, but the initial AOD conditions can affect the amplitude of variation of COT and CWP. Meanwhile, the values of d(CF) are smaller than zero over the ECS. This shows that the cloud fraction in high AOD conditions over ECS decreases less than that in low AOD conditions. However, Meskhidze et al. (2009) found that an increase of the aerosol concentration may lead to enhanced reduction of afternoon cloud coverage and optical thickness for marine stratocumulus regions off the coast of California, Peru, and southern Africa. Therefore, the connection between AOD and variation of cloud cover could be a response to regional-scale changes in aerosol covarying with meteorological conditions. The value of d(CF) is overall positive over the PRD, which indicates that over the PRD in high AOD conditions the cloud cover increases much more than the cloud cover decreases in low AOD conditions. Mauger and Norris (2007) have shown that scenes with large AOD and large cloud fraction experienced greater LTS. As regards CTP, we find that the values of d(CTP) are positive over the BTH and PRD urban cluster, but the values of d(CTP) over the other two regions are not significant. It indicates that in high AOD conditions over the PRD region the CTP increases much more than the CTP decreases in low AOD conditions. We can conclude that the variation in d(Cloud_X) is different for continental and oceanic clouds. This applies to CDR, cloud fraction (CF) and CTP, but not to COT and CWP. Table 2 summarizes the differences between the mean changes in cloud properties for low and high AOD over the timestep of 3 hours.

Based on the above findings, we conclude that over the ECS the values of CDR, CWP and CTP are smaller but the values of COT and CF are larger in high AOD conditions. After the 3 hours timestep, CDR, CF and CTP become smaller, irrespective of the AOD. Furthermore, CDR decreases much more in high AOD conditions but CF and CTP decreases much more in low AOD conditions. In contrast, COT and CWP become larger in both AOD conditions, more significantly in low AOD conditions. Over the urban clusters, COT and CWP also increase over the timestep in both AOD conditions, especially for the low AOD condition. For CF the values in low AOD conditions decrease over the timestep. The CTP change behaves differently among the three urban clusters during the 3 hours."

15. Comments: (15) Page 15, Lines 7: presumably LTS

Answer: Yes, we made this change (see pg.16 line 7).

16. Comments: (16) Page 15, Line 12: I read exactly the opposite, it looks like there is a high impact of aerosol with descending air parcels.

Answer: The effects of initial cloud fraction and meteorological conditions on the change in CF under low and high AOD conditions after the 3 hours timestep over land are also explored. In our new version manuscript, there are two cases are considered: (1) when the cloud cover increases (ΔCloud_X>0); (2) when the cloud cover decreases (ΔCloud_X<0). So, the results and discussions have been changed. We rephrased the sentence in the revised manuscript (see page 16 lines 13-28).

17. Comments: (17) Page 15, Line 18: Is this change a very large relative humidity statistically significant or just noise?

Answer: The effects of initial cloud fraction and meteorological conditions on the change in CF under low and high AOD conditions after the 3 hours timestep over land are also explored. In our new version manuscript, there are two cases are considered: (1) when the cloud cover increases (ΔCloud_X>0); (2) when the cloud cover decreases (ΔCloud_X<0). So, the results and discussions have been changed. We rephrased the sentence in the revised manuscript (see page 17 lines 1-8).

18. Comments: (18) Page 15, Lines 23: LTS is almost always positive

Answer: Yes, we made this change (see page 17 lines 11-12). "A positive LTS is associated with a stable atmosphere in which vertical mixing is prohibited; negative PVV indicates local upward motion of air parcels." has changed to "Low LTS represents an unstable atmosphere and high LTS represents a stable atmosphere."

19. Comments: (19) Page 15, Line 25: 27K is a very high value for LTS and does not distinguish much between high and low values.

Answer: The effects of initial cloud fraction and meteorological conditions on the change in CF under low and high AOD conditions after the 3 hours timestep over land are also explored. In our new version manuscript, there are two cases are considered: (1) when the cloud cover increases (ΔCloud_X>0); (2) when the cloud cover decreases (ΔCloud_X<0). So, the results and discussions have been changed. We rephrased the sentence in the revised manuscript (see page 17 lines 9-16).

20. Comments: (20) Page 16, Line 4: Why is the initial cloud fraction included if its impact is not clear? Can we learn anything from it?

Answer: The effects of initial cloud fraction and meteorological conditions on the change in CF under low and high AOD conditions after the 3 hours timestep over land are also explored. In our new version manuscript, there are two cases are considered: (1) when the cloud cover increases ($\Delta Cloud\_X>0$); (2) when the cloud cover decreases ($\Delta Cloud\_X<0$). So, the results and discussions have been changed. We rephrased the sentence in the revised manuscript (see page 17 lines 17-24).

21. Comments: (21) Page 17, Line 28: This seems like something that could receive more discussion.

Answer: We rephrased the sentence in the revised manuscript (see page 20 lines 7-13). Text was rephrased as follows.

Page 20 lines 6-13: The results show that cloud cover increases much more for high AOD under stronger upward motion of air parcels; Meanwhile, 7 the increase rate of cloud cover is larger for high AOD with increasing RH when RH greater than 20%. 8 With regarded to the effect of LTS on the change of cloud cover, scenes with large cloud fraction 9 variation experience large AOD and large LTS when LTS smaller than 10. Conversely, scenes with 10 smaller cloud fraction variation experience large AOD and large LTS when LTS larger than 10 and 11 smaller than 20. We also find that smaller increase rate of cloud fraction occurs when scenes 12 experience larger AOD and larger initial cloud cover.

22. Comments: (22) Page 17, Line 13: This relationship between initial cloud fraction and changing cloud fraction is mentioned again with very little explanation as to why.

Answer: We made this change (see page 19 lines 28-29). Text was added as: 'Both COT and CWP increase over land and ocean after the timestep, irrespective of the AOD. The variation trend of COT and CWP after 3 hours depends little on the initial AOD, but the initial AOD conditions can affect the amplitude of variation of COT and CWP. '

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Reply to comments on "Satellite-based estimate of the variability of warm cloud properties associated with aerosol and meteorological conditions"

October 30, 2018

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

Comments

1. Comments: (1) Page 4, Line 20: How is the possibility of vertical segregation of cloud and aerosol accounted for? For example, the presence of a lofted aerosol layer in the same scene as the low clouds?

Answer: Caution is warranted in investigating the satellite-derived relations between aerosol and cloud properties. With MODIS we cannot resolve the height of aerosol and cloud layers, or detect aerosol above clouds. However, the physical and optical properties of clouds and aerosol are quite different and these are used to separate aerosols from clouds. These tests will not work for lofted aerosol layers above clouds because the cloud reflectance overwhelms that of aerosols. Some other sensors rather than MODIS have specific instrument characteristics that allow for this separation.

Firstly, as we know, CALIPSO and CloudSat can provide the height of aerosol layer and cloud layer, however, the relatively low number of MODIS-CALIPSO coincidences limits the further binning of the data required to investigate this issue. Secondly, when 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 0.8 were excluded as a rough attempt to exclude cloud-contaminated AOD to reduce the uncertainty in the observed ACI. As reported by Yuan et al. (2010), the potential artefact mentioned above does not seem to be the primary cause for the observed relationship between aerosol and cloud parameters. Further investigations are needed to fully analyze and explain the observed phenomena.

2. Comments: (2) Section 3.2: Should this not include a sentence on how the significance of results is to be determined? The results table speaks of statistical significance but I'm not clear how this is measured.

Answer: Yes, we agree. We made this change in the revised manuscript (see page 6, line 21-23).

Page 6, line 21-23: Text was added as:' Student's t test is used to determine whether two data sets are significantly different from each other. The marker ** at the top right corner of symbol "+" (or "-") denotes that the difference between a change in cloud property and zero is significant (at 95% confidence level).'

3. Comments: Section 4.1: I was under the impression from section 3.1 that the difference in cloud properties at time=0 had been removed by re-sampling the data. I think the relationship between sections 3.1 and 4.1 needs to be developed with the lay reader in mind.

Answer: Normalised histograms of cloud properties for the high and low AOD populations are made for the whole region (Section 3.1), because the data volume based on each 1° x 1° location is relatively small. However, the difference between the cloud properties for low and high AOD at the start time is based on each 1° x 1° location (Section 4.1). So the difference of the cloud properties between the low and high AOD at the start time is not zero. In order to make the reader understand, text was added as follows.

page 5 line 37-39 and page 6 lines 1-2: Text was added as: 'Note that here and in the following sections, normalised histograms of cloud properties for the high and low AOD populations are made for the whole region (Section 3.1), because the data volume based on each 1° x 1° location is relatively small. However, the difference between the cloud properties for low and high AOD at the start time is based on each 1° x 1° location (Section 4.1). So the difference of the cloud properties between the low and high AOD at the start time is not zero.'

Page 9, line 14-16: Text was added as: 'Although normalized histograms of meteorological parameters are made for high and low AOD conditions at the start time, the normalization described in Sect. 3.1 is based on the whole region. Differences in meteorological conditions may still occur between each 1° x 1° grid cell.'

Meanwhile, in order to consider the effect of meteorological conditions on the relationship between aerosol and cloud further, we analyze the meteorology of the different regions in section 4.2 (see page 9-10). This new section 4.2 "The meteorology of the four target regions" reads:

4.2 The meteorology of the four target regions

The meteorological and aerosol effects on clouds are reported to be tightly connected, and this connection must be accounted for in any study of aerosol-cloud interactions (Stevens and Feingold, 2009; Koren et al., 2010). Although normalized histograms of meteorological parameters are made for high and low AOD conditions at the start time, the normalization described in Sect. 3.1 is based on the whole region. Differences in meteorological conditions may still occur between each 1° x 1° grid cell. In this study, we analyze the meteorology of the different regions, in support of the interpretation of the regional variation of the relationships between aerosols and clouds.

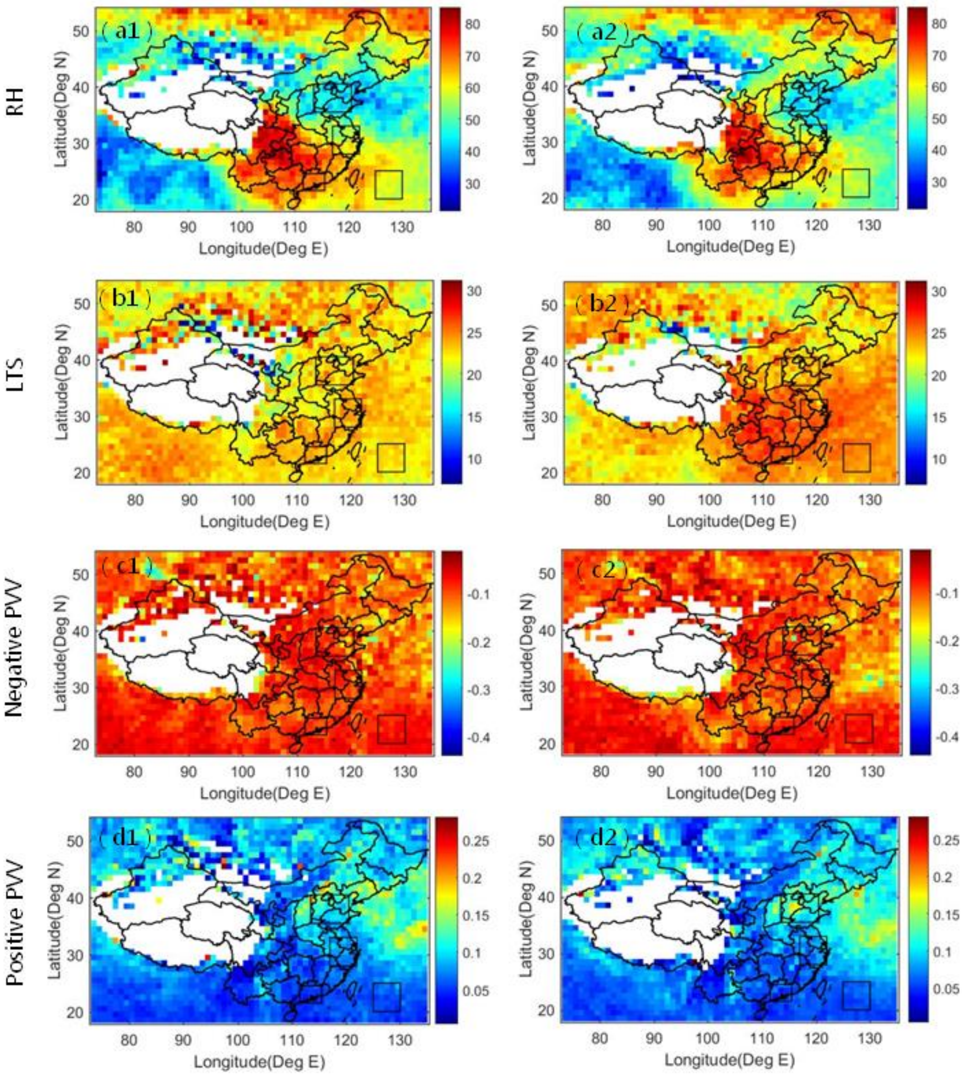

Figure 4 Spatial distributions of meteorological parameters (top to bottom: RH, LTS, positive PVV and negative PVV) at the start time of the timestep (MODIS/Terra) for low AOD conditions (left, a1-d1) and for high AOD conditions (right, a2-d2). All the data are averaged over all years between 2008 and 2017.

The spatial variations of the aerosol and cloud properties over the four regions, averaged over the years 2008-2017, are shown in Fig. 4. Over the urban clusters, we can see an increasing north–south pattern in RH and LTS, with the lowest values found in the PRD. For the negative PVV, the spatial distributions for the low and high AOD situations are remarkably similar, with the highest values over the BTH and decreasing toward the south to near zero over the PRD. In contrast, the positive PVV is smallest over the BTH, with little variation over the study area. Overall, the meteorological parameters over the YRD and PRD are similar to those over the ECS, irrespective of the AOD. Furthermore, the LTS is significant larger in the high AOD conditions for all the four regions. Zhao et al. (2006) proposed that the enhancement in the atmospheric stability tends to depress upward motion and precipitation, leading to an increase in aerosol particles. The spatial distributions of both positive and negative PVV in the low AOD conditions are similar to those in the high AOD conditions.

4. Comments: Page 9, Line 31: I'm not sure what this sentence means – either the ECS is your marine study area or it isn't. I would caution against using parentheses in the way used in this sentence, because it is well - recognised to make the sentence much harder to comprehend whilst reading. The writer should be aiming to help the reader assimilate the information at normal reading speed, not to slow them down with internal opposites that require going back and forth over the sentence repeatedly.

Answer: Following reviewers' comments, we use collection 6.1 data and reanalyze all the data for the whole acquisition period between 2008 and 2017, rather than collection 5.1 data from 2008 to 2011. As a result, the data base was expanded and provides more cases. We have included this information throughout the revised manuscript (all the figures were changed/modified in this respect). So, we reorganize the sentences in the section (see page 11, line 16-17) and do not use parentheses in the way in this sentence throughout the revised manuscript.

"Over the ECS, in low AOD conditions, CDR decreases during the timestep while COT and CWP increase (Figure 4). For high AOD conditions, the variations of the cloud properties (CDR, COT and CWP) during the timestep are similar to those for low AOD conditions (Figure 5). Furthermore, it appears that COT and CWP increase more at low AOD than at high AOD. Having a closer look at the CF/CTP variation in both low and high conditions over ocean, we can find that CF decreases (CTP increases) in low AOD conditions and CF increases (CTP decreases) in high AOD conditions over ocean, albeit not over ECS. " has been changed to "Over the ECS, in both low and high AOD conditions, CDR, CF and CTP decrease during the timestep while COT and CWP increase (see Figure 5). " in the revised manuscript (see page 10). Also, as we merged section 4.2 with section 4.3 (as new Section 4.3, see pages 10-15), more discussion has been shown in section 4.3 of the revised manuscript (see page 14-15).

5. Comments: Page 10, Line 1: is the other "significant difference" one or two differences? The sentence seems to suggest that CF and CTP co-vary, which in turn suggests they need not have been studies separately.

Answer: As mentioned above, due to the larger data set, the variation of cloud properties to the aerosol environment has become more clear. This is shown throughout the revised manuscript (all the figures were changed/modified in this respect). We reorganized the sentences in the section (see page 11, line 19-28).

The paragraph "In general, the variations in cloud properties over land are similar to those over ocean for both low and high AOD conditions over 3 hours. Two significant differences are found between land and ocean areas. One is that CDR increases over land but decreases over ocean after the timestep, another significant difference is that CF decreases (CTP increases) for low AOD condition but CF increases (CTP decreases) for high AOD condition over ocean after the timestep, whereas CF increases (CTP decreases) for both low and high AOD conditions over land after the timestep. We can conclude that the variation of cloud properties after 3 hours depends little on the initial AOD over land, even though differences exist among the urban clusters. The increase in afternoon cloud fraction over land is consistent with previous studies concluding that continental warm clouds are likely to be well developed (Wang et al., 2014; Kourtidis et al., 2015). The decrease in afternoon cloud cover over ocean confirms that the largest cover for marine clouds is reached early in the morning (Meskhidze et al., 2009). Table 2 summaries the differences in cloud properties between the Aqua and Terra overpasses for high and low AOD conditions over land and ocean during the time period 2008-2011, respectively." **has been changed to** "In general, the variations over 3 hours in COT and CWP over land are similar to those over ocean for both low and high AOD conditions. Another significant similarity is that CF decreases for low AOD conditions over land and ocean during the 3h timestep. Having a closer look at the CF variation over the YRD and PRD, we see that CF increases in high AOD conditions during the 3h timestep. This implies that the variation of CF may depend on the initial AOD conditions. The decrease in afternoon cloud cover over ocean confirms that the largest cover for marine clouds is reached early in the morning as was also concluded by Meskhidze et al. (2009). Meanwhile, a significant difference is found between land and ocean areas, i.e. in high AOD conditions CDR increases over land but decreases over ocean during the 3h timestep. Table 2 summaries the differences in cloud properties between the Aqua and Terra overpasses for high and low AOD conditions over land and ocean during the time period 2008-2017."

6. Comments: Figures 3-5: might it be possible to shade the graphs on the right-hand columns of these figures to show when changes are self-consistent from a microphysics or cloud dynamics point of view?

Answer: We do not understand what the reviewer means with "self-consistent from a micro-physics or cloud dynamics point of view". Hence, we have not added a shading to these graphs.

7. Comments: Section 4.3 and Figure 6: This reader is left feeling that there is very little added value in this section. The statistics all look close to zero and noisy and the text doesn't make any very strong statements over and above those from previous sections. Is this section really necessary?

Answer: As mentioned above, in response to the reviewers' comments, we use collection 6.1 data and reanalyze all the data for the whole acquisition period between 2008 and 2017, rather than collection 5.1 data from 2008 to 2011. Therefore, the variation of cloud properties to the aerosol environment has become more clear. This is explained throughout the revised manuscript (all the figures were changed/modified in this respect). Furthermore, Section 4.2 was merged with section 4.3 (as new Section 4.3, see pages 10-15), explicitly examining the difference of cloud properties in relation to aerosol environment. Also a new Section 4.2 was added (see response to question 3) to describe the meteorology of the four target regions, in support of the interpretation of the regional variation of relationship between aerosol and cloud (see page 9-10).

8. Comments: Section 4.4 is too definitive considering the uncertainty shown in Figure 7. I also missed a tie-back to basic cloud physics – how is the reader to interpret the effect of aerosol concentration on cloud parameters when the air is descending and cloud formation therefore suppressed? Some context is required here to help the reader who is not familiar with such analyses.

Answer: Yes, we made this change (see page 16 lines 13-16). "The presence of upward motion, as indicated by negative PVV, can enhance the interaction between aerosol particles and clouds as it makes the ambient environment favorable for cloud formation, and vice versa (Jones et al., 2009)." has been changed to "The presence of upward motion, as indicated by negative PVV, can enhance the interaction between aerosol particles and clouds as it promotes vertical mixing of the aerosol particles and thus reach the cloud condensation level where they grow into cloud droplets (Jones et al., 2009)." As the variation of cloud properties to the aerosol environment has become more clear, we also reorganized the sentences in the Section 4.4 (see pages 16-17).

Minor comments

1. Comments: (1) Page 2, line 36: delete 'desperately'

Answer: Yes, we made this change (see page 3, line 1).

2. Comments: (2) Page 6, Figure 2: would plotting on log axes help the differences to be visible? For the caption: presumably this is an example of a PDF of CF?

Answer: We use collection 6.1 data and reanalyze all the data for the whole acquisition period between 2008 and 2017, rather than collection 5.1 data from 2008 to 2011. So, Figure 2 has also been changed and the differences can be seen easily (see page 6).

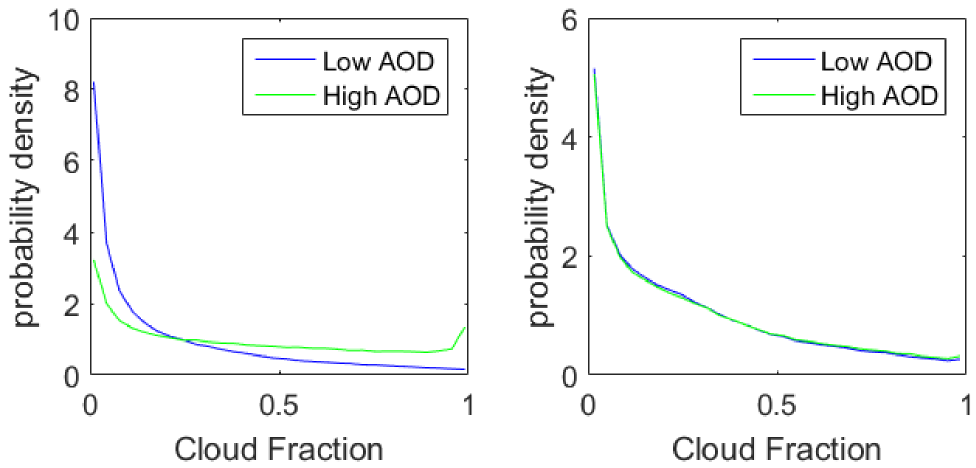

Figure 2. An example of the probability density distribution of warm cloud fraction (CF) for low and high AOD conditions. (a) there is a strong link between AOD and CF before histogram normalization, (b) the link is reduced after histogram normalization.

3. Comments: (3) Page 6, line 12: "Cloud_X (where X=CF, COT, CWP, CDR or CTP)" – just for extra clarity.

Answer: Yes, we made this change (see page 6, line 11).

4. Comments: (4) Page 6, line 13-16: "_Cloud_X[High AOD]" should be overbarred in text as in equation.

Answer: Yes, we made this change (see page 6, line 12).

5. Comments: (5) Page 9, line 27: plot should be plotted.

Answer: We use collection 6.1 data and reanalyze all the data for the whole acquisition period between 2008 and 2017, rather than collection 5.1 data from 2008 to 2011. So, Figure 4 and 5 have been changed to Figure 5 and 6 (see page 12-13).

6. Comments: (6) Page 15, line 12: "when PVV is positive" is more consistent.

Answer: Because the analyzed dataset was different (MODIS C6.1 versus older versions), the result also changed. We rephrased the sentence in the revised manuscript (see page 16).

7. Comments: (7) Page 17, line 1: the statistical methods are not described anywhere that I can see.

Answer: Page 19, lines 8-9: 'Data over these four study areas were collected for the years 2008 to 2017, and analyzed using statistical methods.' was changed to ' Data over these four study areas were collected for the years 2008 to 2017 and analyzed in statistical sense.' Here, we mean with statistical sense that we looked at the 10-year mean properties rather than at individual case studies.

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