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Atmospheric Chemistry and Physics Discussions

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 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 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). The effects of initial cloud fraction and meteorological conditions on the change in CF are also explored, showing that 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 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). 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 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). 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, 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 3. The results and analysis are presented in section 4. Overall conclusions and potential future improvements are discussed in 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 in eastern China 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 often impacted by aerosol particles transported from the highly industrialized eastern China (Wang et al., 2014). The study period is 4 years, i.e. 2008-2011.

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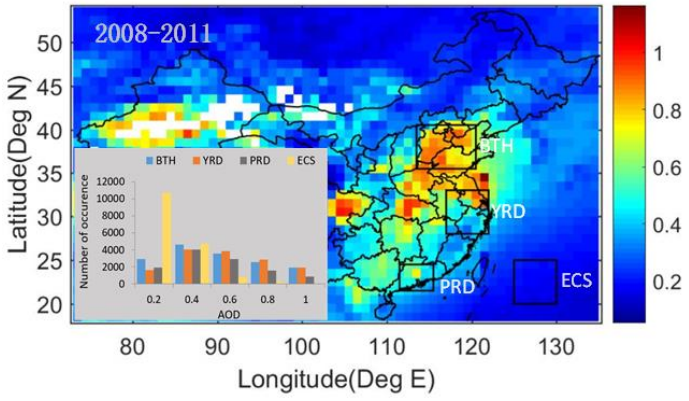

Figure 1. Map of MODIS/AQUA level 3 AOD over Eastern China averaged over the period from 2008 to 2011. The location of the four clusters (three urban and one ocean) studied here (Beijing-Tianjin-Hebei: BTH, Yangtze River Delta: YRD, Pearl River Delta: PRD and Eastern China Sea: ECS) are marked with black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

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 (Levy et al., 2010; Xiong et al., 2008). The MODIS L3 collection 5.1 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 than 700 hPa, CTT greater 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 is representative of typical thermodynamic conditions (Klein and Hartmann, 1993). 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 data samples are available for the 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. After that, both conditions have almost the same CF distribution at the start of the timestep through the normalization process, 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 CDR, COT, CWP and CTP at

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the start time, even though we find that the normalization for the cloud fraction made the biggest difference by far. In the further analysis, we only take a subset of original data by removing random samples until the histograms are similar.

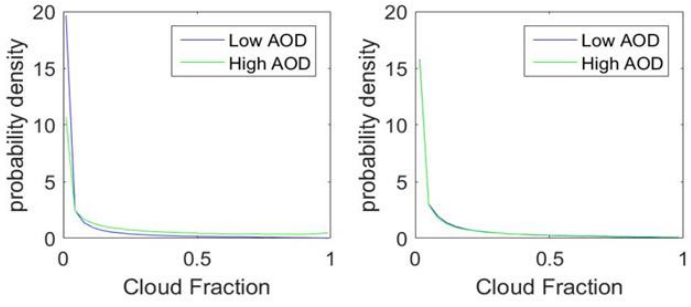

Figure 2 Probability density distribution of warm cloud fraction (CF) for low and high AOD conditions. (a) there is a strong link between AOD and CF before histogram normalization, (b) the link is reduced after histogram normalization.

3.2 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 of the relationship between AOD and cloud parameters over the timestep. For cloud property Cloud_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 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.

4 Results and Discussions

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 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 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 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). 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 higher at high AOD over the ECS 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. Figure 3(c1-c2) shows that CWP is lower at high AOD over the ECS, which is not in agreement with COT variation. In contrast, over the PRD, CWP behaves similar to COT and is higher at high AOD. 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. Meanwhile, over the YRD urban cluster CF is lower and CTP is higher at high AOD. In contrast, over the BTH 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.

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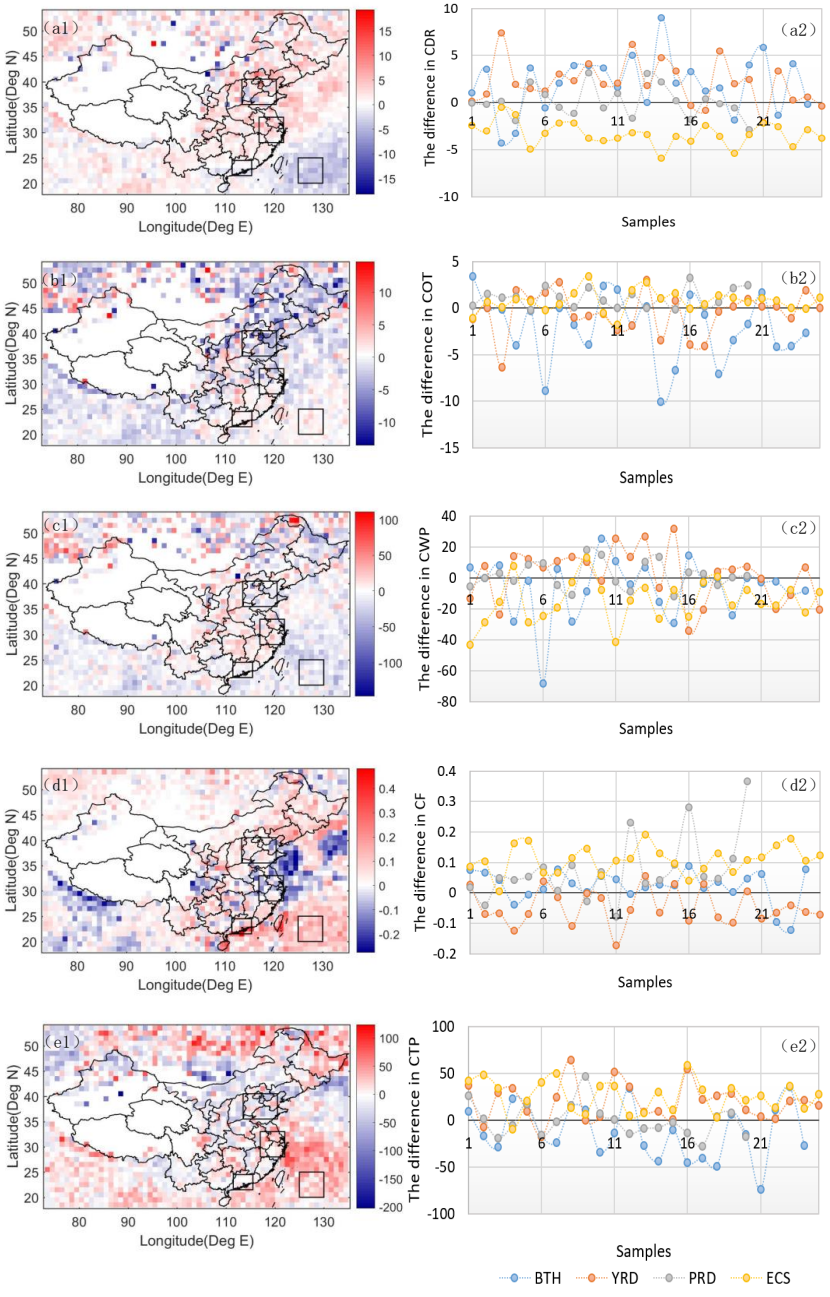

Figure 3 Spatial distribution of the differences in cloud properties (top to bottom: CDR, COT, CWP, CF and CTP) between the highest and the lowest MODIS AOD quartiles (highest - lowest) at the start time of the timestep (MODIS/Terra) (left, a1-e1) and sample series of the differences in cloud properties (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.

To better characterize the variation in cloud properties between high and low AOD, Table 1 summarizes the

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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. 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 predictable over land (BTH, YRD and PRD) than over ocean (ECS).

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 '~' indicates the response of cloud properties to the increasing AOD is not significant.

4.2 The mean change in cloud properties over the timestep for low and high AOD

The mean afternoon values of cloud properties 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 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 5 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 4 years. Concerning the effect of aerosol loading on cloud parameters in each urban cluster, 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, but for the variation of CTP, a significant decrease occurs over the YRD for both low and high AOD conditions. By contrast, increases of CDR, COT and CWP were observed for both low and high AOD conditions over the PRD urban cluster, which is statistically significant. From the perspective of considering all urban clusters (BTH, YRD and PRD), all studied cloud properties (CDR, COT, CWP and CF) increase except CTP over land, which decreases during the timestep, for both low and high AOD (see red samples plot in Figures 4 and 5).

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.

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

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that CDR increases over land but decrease 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.

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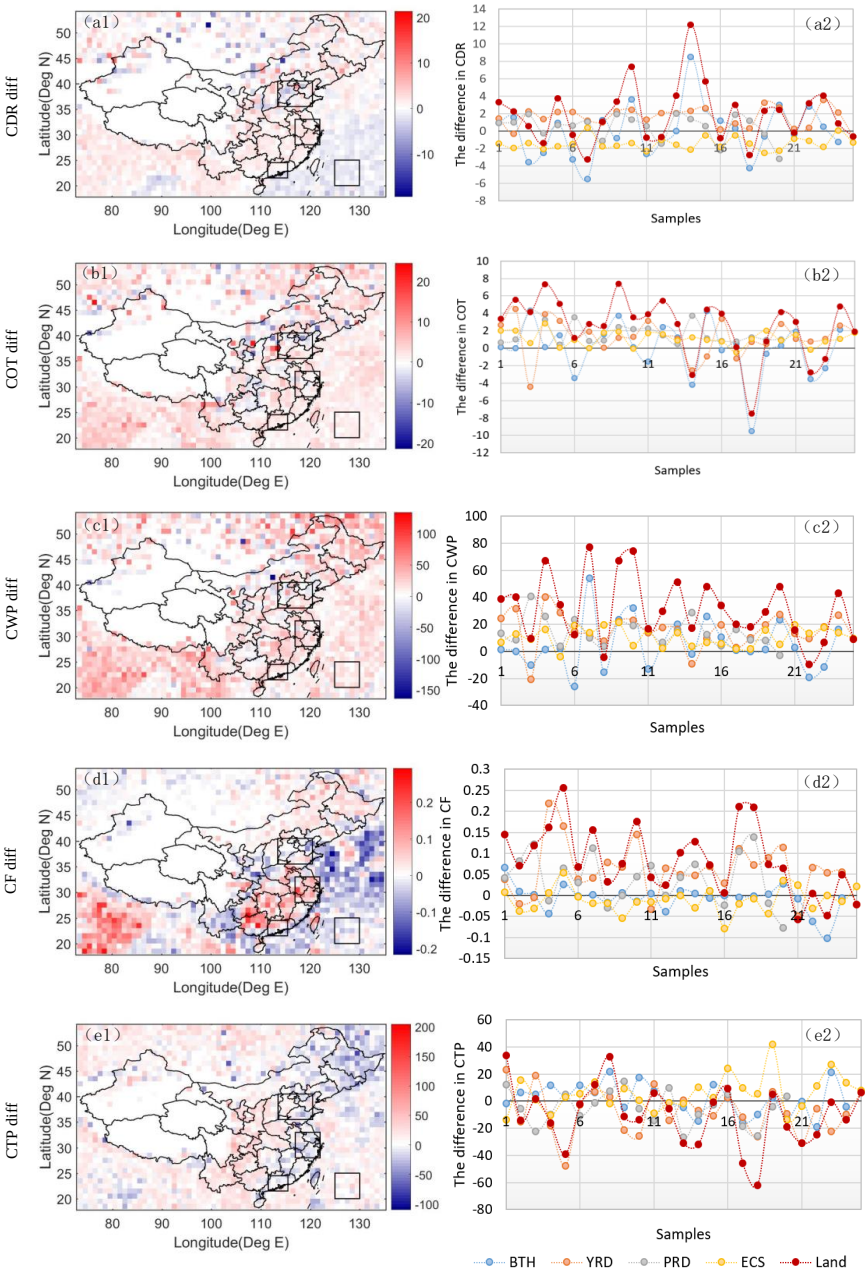

Figure 4 Spatial distributions of differences in cloud properties (CDR, COT, CWP, CF and CTP) between Aqua and Terra overpasses (3 hours) for the lowest MODIS/Terra AOD quartiles (left, a1-e1). Sample series of the differences in cloud properties (CDR, COT, CWP, CF and CTP) between the values at the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

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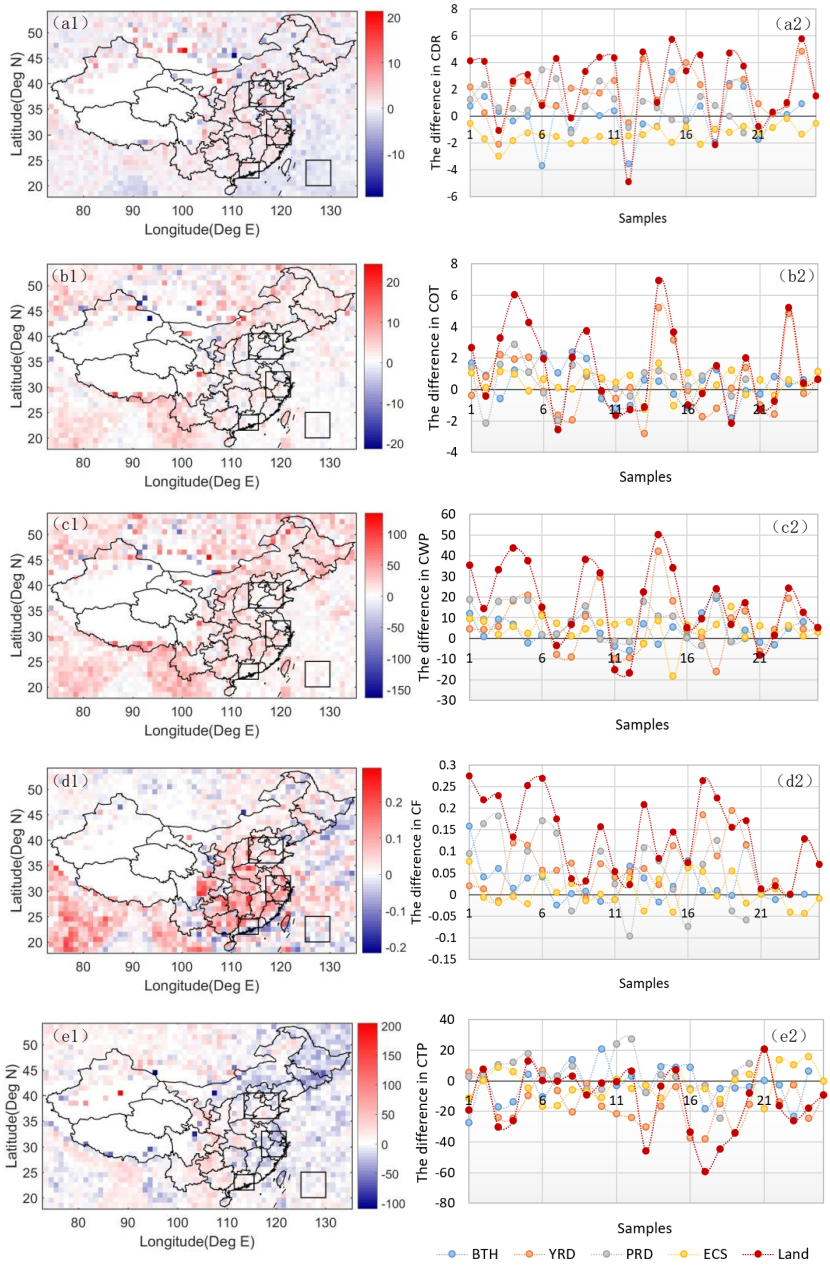

Figure 5 Spatial distributions of differences in cloud properties (CDR, COT, CWP, CF and CTP) between Aqua and Terra overpasses (3 hours) for the highest MODIS/Terra AOD quartiles (left, a1-e1). Sample series of the differences in cloud properties (CDR, COT, CWP, CF and CTP) between the values at the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

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

	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 3.2) are investigated to identify the effect of aerosol particles on the cloud properties. Figure 6 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 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. Over the ECS the values of d(CDR) also vary around zero, which indicates that the CDR of high AOD decreases as much as that of low AOD over ocean. Most of the d(COT) values are negative over the four regions, especially for the PRD and ECS. This shows that the COT increases less in high AOD conditions than in low AOD conditions, over both land and ocean. Likewise, the values of d(CWP) are almost all negative over the four regions although over the BTH urban cluster the values are less 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 at low AOD conditions. Meanwhile, the values of d(CF) are larger than zero over the BTH urban cluster. This shows that the cloud fraction in high AOD conditions over BTH increases much more than that in low AOD conditions. The value of d(CF) is overall positive over land, which indicates that over land in high AOD conditions the cloud cover increases much more than 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. As regards CTP, we find that the values of d(CTP) are positive over the PRD urban cluster, but negative over the YRD and ECS regions. 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 difference between the mean changes in cloud properties for low and high AOD over the timestep of 3 hours.

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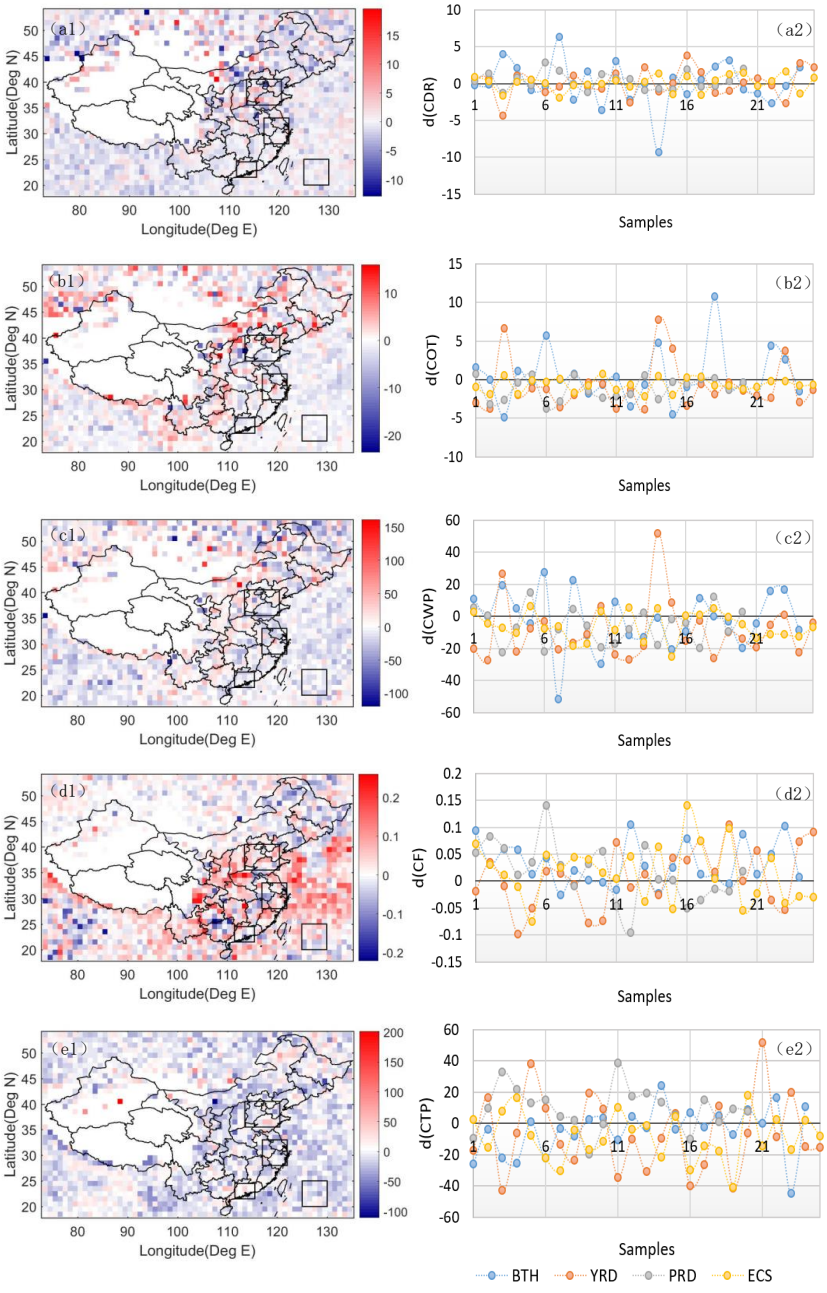

Figure 6 Spatial distributions (left, a1-e1) and sample time series (right, a2-e2) of d(Cloud_X) (as defined in sect. 3.2) for CDR, COT, CWP, CF and CTP over Eastern China during the time period 2008-2011.

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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 meteorological parameters, respectively. Figure 7 shows the effect of meteorological factors (PVV, RH, LST and initial cloud fraction) on the d(CF) 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 makes the ambient environment favorable for cloud formation, and vice versa (Jones et al., 2009). Figure 7(a) shows that the d(CF) increases with the PVV when the PVV is negative. In contrast, the d(CF) decreases with the PVV when the PVV is larger than zero. This indicates that the upward motion of air parcels can enhance the cloud cover much more in conditions with high AOD than in conditions with low AOD. 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(b) shows that the d(CF) decreases with increasing RH when RH is lower than 92%. This implies that the cloud cover increases much more in low AOD environment than in high AOD environment with increasing RH. However, when RH is larger than 92%, a strong increase of d(CF) occurs due to activation of CCN and formation of clouds. It should be noted that the variation of d(CF) with increasing RH is uncertain as the sample sizes of high and low AOD conditions are small. 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. Figure 7(c) shows that the d(CF) increases with increasing LTS when LTS is lower than 27, but decreases with increasing LTS for higher values. 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

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environment with high AOD concentrations. However, the sample sizes of high and low AOD conditions are small when LTS is higher than 27. Therefore, the relationship between d(CF) and LTS is uncertain. Figure 7(d) shows that there is weak negative relationship between d(CF) and initial cloud fraction. The effect of initial cloud fraction on d(CF) is not clear.

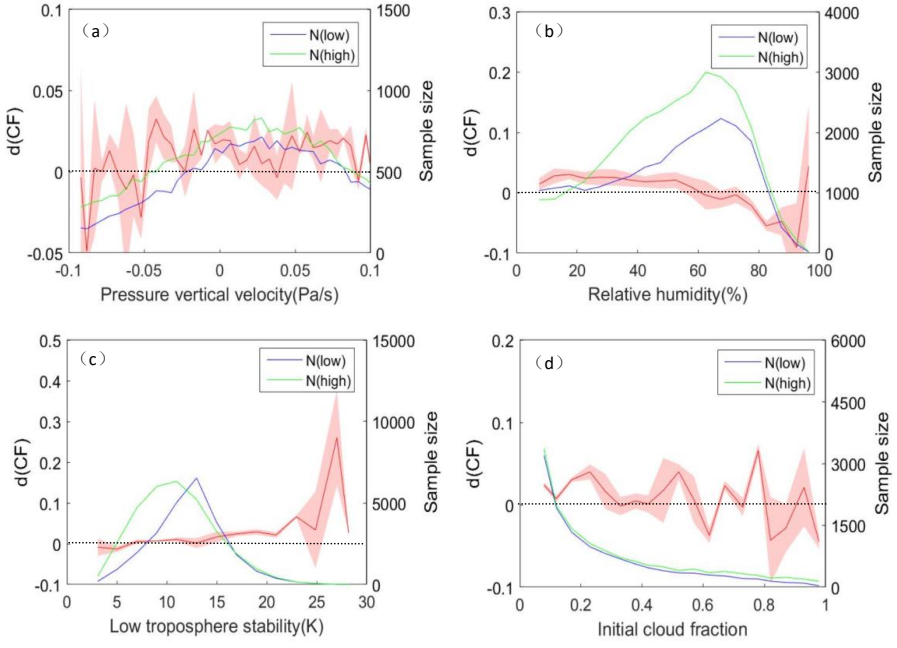

Figure 7 Variation of d(CF) (red) as function of initial meteorological parameters and cloud fraction for warm clouds over land. The distribution of points for low (blue) and high (green) AOD as a function of meteorological parameters is shown by the solid lines. This plot is composed from MODIS data (including Terra and Aqua) for all warm cloud points over the years 2008-2011. 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.

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, as indicated by AOD, 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, and analyzed using statistical methods. Both MODIS/Terra and MODIS/Aqua data were used to study the difference of cloud properties between the morning and early afternoon, i.e. with a time difference of 3 hours.

In order to reduce the difference of the initial conditions between high and low AOD conditions at the start of the timestep, normalized histograms of 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 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 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.

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. The results show that 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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