Effects of Shipping-originated Aerosols on Physical Cloud Properties over Marine Areas near East China

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ABSTRACT

Shipping emissions have received increasing attention due to their influence on regional climate, and air quality at ports and in coastal areas around the world. In the context of climatology, the effects of shipping-originated aerosols (using the aerosol optical depth, AOD_{ship} , as a proxy) on clouds in marine areas near East China were examined based on multi-satellite datasets. On average, AOD_{ship} is approximately 0.17, 0.20, 0.15 and 0.13 in the different seasons, contributing 23%, 30%, 36% and 25% of the total AOD, respectively. In remote sea areas, AOD_{ship} is generally higher in spring and summer and lower in autumn. Statistical analysis shows that it is strongly related to the cloud parameters, such as the cloud fraction (CF), cloud optical thickness (COT) and cloud effective radius (CER) in the liquid phase. In particular, CER and AOD_{ship} exhibit a positive correlation in winter but a weakly negative correlation in summer over the northern East China Sea and a positive correlation in spring and summer over the Yellow Sea. In all four seasons, COT and CF decrease as AOD_{ship} increases. The correlations between AOD_{ship} and the cloud properties are stronger ($R > 0.3$) when the aerosol-cloud layers are well mixed than when they are separated, indicating that shipping-originated aerosols drawn into the cloud body can directly affect the microphysical properties of cloud droplets during cloud formation. The water vapor content and upward air motion are key thermodynamic conditions within the low atmospheric layers under the cloud bottom that play an important role in cloud formation and development. Our results provide new insight into the influence of shipping emissions on clouds in Asian marine areas.

Keywords: Shipping emissions; Aerosol; Cloud property; East China.

INTRODUCTION

Aerosols' direct and indirect effects can disturb the earth's climate system (IPCC, 2014). The effects of aerosols on clouds has become the largest uncertainty in assessing the anthropogenic aerosols' contribution to global climate change (Twomey, 1974; Rosenfeld, 2000; Kaufman *et al.*, 2012; Gryspeerd *et al.*, 2014). As some aerosols act as cloud condensation nuclei (CCN), an increase in particles entrained into clouds can lead to more and smaller cloud

droplets under constant liquid water path (Twomey, 1977) and then result in cloud albedo growth (Twomey, 1977). Meanwhile, the decrease of cloud droplet size causing clouds living longer with more pollution can impact precipitation cycles (Albrecht, 1989). Additionally, a possible reduction in precipitation frequency of liquid clouds through the suppression of drizzling prolongs the cloud lifetime (Albrecht, 1989).

Marine transportation has been viewed as one of the largest, least regulated sources of anthropogenic pollutants over oceans and coastal areas (Marmer and Langmann, 2005). Ships usually release massive amounts of primary pollutants, such as particles, NO_2 , CO, NMVOCs and SO_2 , which results in significant local and spatial air pollution in ports and harbors, as well as secondary particles (e.g., secondary organic aerosol (SOA)) that are potentially harmful to human health (Saxe and Larsen, 2004). The

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ship exhaust amounts approximately 400 km within the coastline of the Yangtze River Delta (YRD) bordering the East China Sea were estimated to be approximately 3.8×10^5 , 7.1×10^5 and 5.1×10^4 ton y^{-1} annually for SO_2 , NO_x and $PM_{2.5}$, respectively (Fan *et al.*, 2016). Shipping emissions are a main contributor to NO_2 and SO_2 over many marine areas of the world, accounting for 0.5–5% of all NO_2 and SO_2 emissions at most ports and, in some ports, exceeding 10% (Dalsøren *et al.*, 2009). Furthermore, 11% of nitrate wet deposition and 4.5% of sulfur wet deposition globally result from shipping emissions, whereas in certain coastal regions up to 15–50% of nitrate and sulfur wet deposition result from shipping emissions (Dalsøren *et al.*, 2009).

The influence of rising shipping pollution on clouds and precipitation has gradually become an open question (Rosenfeld *et al.*, 2008; Stevens and Feingold, 2009). In recent years, some studies have explored the effect of shipping-originated aerosols on marine clouds (Petzold *et al.*, 2008, 2010; Coggon *et al.*, 2012; Juwono *et al.*, 2013; Russell *et al.*, 2013). Even low concentrations of larger CCN originated from ships can decrease cloud albedo by enhancing collision-coalescence, as proposed by Feingold *et al.* (1999). Wang *et al.* (2014a) figured out the effects of emissions from shipping on stratocumulus cloud water elemental composition during the 2011 E-PEACE field campaign. Chen *et al.* (2012) found occurrence of lower cloud albedo in ship tracks. The droplet sizes in clouds over ship tracks decrease and the total number of cloud droplets substantially increase compared to those in adjacent clouds, as observed by Radke *et al.* (1989). Sorooshian *et al.* (2015) found that aerosol particle concentrations behind ship tracks exceed those specified in the North American Emission Control Area (ECA; a sulfur content of marine fuel of no more than 1%), and their number enhancement ratios decrease with growing downwind distance and altitude. Cloud droplet number has been reported to increase with aerosol loading over the East China Sea (Bennartz *et al.*, 2011). Significant negative correlation coefficients were found between aerosol optical depth (AOD) and cloud effective radius (CER) for lower AOD levels over the East China Sea (Tang *et al.*, 2014); meanwhile, positive correlations exist for higher AOD levels. In contrast, significant positive correlations exist over the Yellow Sea (YS) between the Korean Peninsula and Chinese mainland.

Container throughput is an important indicator to reflect the competitiveness of the shipping industry. By the end of 2016, there were 170 ultra-large ships with over 10,000 TEU on daily Asia-Europe routes, accounting for 43% of total ships (Tan, 2017). The East China region has densely clustered ports, and 4 of the top 10 ports in the world are located there, including the ports of Shanghai, Ningbo-Zhoushan, Qingdao and Suzhou. Meanwhile, the cargo throughput of these ports has experienced a rapid increase in recent years. To date, few studies have focused on the effect of aerosols from shipping-originated emissions on local air quality and regional climate over marine areas near East China.

In this study, the effect of shipping-originated aerosols

on clouds over marine areas near East China was examined on the basis of multi-satellite data. The purpose is to exhibit the spatial distribution and seasonality of shipping-originated aerosol optical depth (AOD_{ship}) over the focused domain and to evaluate the relationships between AOD_{ship} and main macro- and micro-scale cloud physical properties. Moreover, the relevant factors to influence the above correlations were also analyzed, including aerosol-cloud mixture and meteorological conditions within low atmospheric layers.

DATA AND METHODOLOGY

Study Area

Fig. 1(a) shows the focused domain covering the northern areas of the East China Sea (NECS) and most areas of the Yellow Sea near East China, with an extent of 28–36.5°N and 121–128.5°E. Fig. 1(b) shows the spatial distribution of annual mean primary $PM_{2.5}$ from shipping emissions in 2014 (Chen *et al.*, 2017). In 2014, the estimated shipping emissions of $PM_{2.5}$ in the YRD region (latitude: 28–33.5°N, longitude: 119.5–124°E) were 4.91×10^4 t, accounting for 28–30% of the total shipping emissions over the Bohai Sea (BSA), the YRD and the Pearl River Delta (PRD) (latitude: 21.5–23.8°N, longitude: 112–115.5°E) (Chen *et al.*, 2017). In this study, we mainly discuss possible effects of shipping-originated aerosols on marine clouds over East China Sea areas using various space-based measurements made in 2014.

Remote Sensing Datasets

MODIS

The Moderate Resolution Imaging Spectroradiometer (MODIS) sensor is aboard the Terra and Aqua satellites (Levy *et al.*, 2007). Aerosol products are derived from MODIS data, among which AOD is retrieved using a dark target algorithm or deep blue algorithm (Remer *et al.*, 2005). The standard MODIS Collection 4 Level 2 AOD and FMF (Optical_Depth_Ratio_Small_Ocean) data product was used for identifying the ship aerosols and sea salt aerosols in the study sea area. The standard MODIS Collection 8 Level 3 daily data product at a spatial resolution of $1^\circ \times 1^\circ$ was used for obtaining the spatial distribution of AOD_{ship} and the analysis of the relationships between AOD_{ship} and cloud properties without distinguishing aerosol-cloud overlap, in combination with that from Terra at 10:30 LT and that from Aqua at 13:30 LT (<https://ladsweb.modaps.eosdis.nasa.gov>). Moreover, the corresponding cloud properties were used to match with AOD, including cloud fraction (CF), cloud effective radius in the liquid phase (CER), cloud optical thickness (COT) and cloud water path in the liquid phase (LWP).

CALIPSO and CloudSat

Along with the Aqua satellite, CloudSat, a cloud-observing satellite, and Infrared Pathfinder Satellite Observation (CALIPSO) currently fly in the “A-train” satellite constellation (Stephens *et al.*, 2002). The Cloud Profiling Radar (CPR) sensor is aboard CloudSat to capture

Fig. 1. Schematic diagram of (a) the study area and locations of giant ports, and (b) the spatial distribution of estimated annual PM_{2.5} emissions from shipping in 2014 (Chen *et al.*, 2017).

the vertical structure information of clouds (especially optically thick clouds) (Im *et al.*, 2005; Wang *et al.*, 2013). Thus, CloudSat is more comprehensive in capturing the vertical structure information of clouds. In this study, we utilized the datasets of RO_liq_effective_radius and RO_liq_water_content (2B-CWC-RO.P_R04) and MODIS_Cloud_Fraction_2B_GEOPROF (2B-GEOPROF.P_R05), which are the CloudSat standard data products with a vertical resolution of 480 m and horizontal resolutions of approximately 1.4×1.8 km (<http://www.cloudsat.cira.colostate.edu>).

CALIOP (Cloud Aerosol Lidar with Orthogonal Polarization (CALIOP) on board the CALIPSO is able to detect aerosol and cloud information (Winker *et al.*, 2003). CPR usually miss the optically thin clouds, while CALIOP is sensitive to that (Wang *et al.*, 2013). In this study, we used the CALIOP Level 2 Cloud Layer product and CALIOP Level 2 Vertical Feature Mask product (<https://www-calipso.larc.nasa.gov>). The vertical resolution of the CALIOP profile product is 60 m from a height of -0.5 km to 20.2 km, and the horizontal resolution is 5 km (Liu *et al.*, 2009). The vertical resolution of the CALIOP Vertical Feature Mask product is 30 m from -0.5 km to 8.2 km, 60 m from 8.2 km to 20.2 km, and 180 m from 20.2 km to 30.1 km. The spatial resolution of the CALIOP Vertical Feature Mask product is 333 m from -0.5 km to 8.2 km, 1000 m from 8.2 km to 20.2 km, and 1667 m from 20.2 km to 30.1 km. Combining CloudSat and CALIPSO observations provides new insights into studying the vertical structure and physical properties of clouds (Matrosov, 2007).

Meteorological Data

Meteorological condition and aerosol effects on cloud physical properties are closely related (Stevens and Feingold, 2009). Relative humidity (RH) and pressure vertical

velocity (PVV) that indicates a vertical movement of air mass can affect the relationship between aerosols and clouds (Small *et al.*, 2011; Gryspeerd *et al.*, 2014). The daily wind speed (WS), wind direction (WD) and RH at 950 hPa and the PVV at 750 hPa were obtained from the Final Analysis (FNL) of the National Centers for Environmental Prediction (NCEP). The daily FNL data contains global meteorological fields with $1^\circ \times 1^\circ$ grid horizontal resolution and 26 levels of vertical resolution (10–1000 hPa) every 6 h. Detailed dataset information is shown in Table 1.

Methodology

To maximize the amount of AOD data during targeted periods, we regarded available values observed by either morning or afternoon satellites as the daily AOD of the individual day. When both morning and afternoon measurements were available, their arithmetic average was used as the daily AOD. Otherwise, when they both were unavailable, the daily AOD was viewed as null.

Firstly, the sum of the shipping-originated aerosol AOD and sea salt AOD ($AOD_{\text{ship}} + AOD_{\text{ss}}$) for the study area was screened out based on the FNL data of wind speed and wind direction. In the case that the u component of wind at the 02 area is less than 0 and the v component of wind at the 01 area is greater than 0, whatever the wind speed is, the aerosols are believed to originate from shipping emissions and sea salt, and they contribute mainly to AOD. When the u component of wind at the 02 area is greater than 0 and less than 2.5 m s^{-1} (Nilsson *et al.*, 2001) and the v component of wind at the 01 area is greater than -2.5 m s^{-1} (Nilsson *et al.*, 2001) and less than 0, under which the effect of continental aerosols on total aerosol loading is ignorable and can be excluded, the shipping-originated aerosols and sea salt are also considered as the major contributor to AOD. As wind blows almost from the sea in other areas

Table 1. MODIS, CALIPSO, CloudSat and FNL products used to characterize aerosol and cloud properties.

Data source	Product	Dataset
MODIS	Aerosol (Level 3)	Aerosol_Optical_Depth_Land_Ocean
	Aerosol (Level 2)	Optical_Depth_Land_And_Ocean
		Optical_Depth_Ratio_Small_Ocean
	Cloud (Level 3)	Cloud_Effective_Radius_Liquid
		Cloud_Water_Path_Liquid
CALIPSO	Aerosol (CPro Level 2)	Cloud_Fraction
	Cloud (CPro Level 2)	Cloud_Optical_Thickness_Liquid
	Vertical Feature Mask (Level 2)	Column_Optical_Depth_Aerosols_532
		Column_Optical_Depth_Cloud_532
CloudSat		Feature_Classification_Flags
	Cloud (2B-GEOPROF.P_R05)	MODIS_Cloud_Fraction_2B_GEOPROF
	Cloud (2B-CWC-RO.P_R04)	RO_liq_effective_radius
		RO_liq_water_content
NCEP	FNL	Wind speed (950 hPa)
		Wind direction (950 hPa)
		Relative humidity (950 hPa)
		Pressure vertical velocity (750 hPa)

(excluding 01 and 02 areas), AOD is totally dominated by sea salt aerosols or shipping-originated aerosols. Secondly, based on the criteria for aerosol classification proposed by He *et al.* (2012), $0 < \text{AOD} < 0.5$ and $0.25 \leq \text{FMF} < 0.7$ were selected as criteria for identifying ship and sea salt aerosols (excluding other aerosols) in the study area. The combination of the two methods can effectively exclude the influence of long-distance transport aerosols (dust, continental) and can more accurately identify ship and sea salt aerosols. The statistical results of screened AOD_{ship} plus AOD_{ss} over the study area are shown in Table 2. According to previous research (Kaskaoutis *et al.*, 2007), when $0 < \text{AOD} < 0.2$ and $0 \leq \text{FMF} < 0.7$, the aerosol type is classified as clean ocean aerosol (AOD_{ss}), and the spatio-temporal distributions of AOD_{ss} have little change. In this study, we chose 0.15 as the background AOD_{ss} (Fan *et al.*, 2011), and then we roughly calculated AOD_{ship} by subtracting the background AOD_{ss} from $\text{AOD}_{\text{ship}+\text{ss}}$ that was greater than 0.15. If $\text{AOD}_{\text{ship}+\text{ss}}$ was less than 0.15, AOD_{ship} was regarded as 0. Finally, the spatial distributions of AOD_{ship} in spring (MAM), summer (JJA), autumn (SON) and winter (DJF) were achieved by the above method.

Referring to the study of Costantino and Bréon (2013), in the case of the vertical distance between the bottom of the aerosol layer and the top of the cloud layer was smaller than 100 m or the vertical distance between the top of the aerosol layer and the bottom of the cloud layer was smaller than 100 m, aerosol and cloud layers were considered to be physically interacting (well mixed) (Fig. 2(a)). When their vertical distance exceeded 750 m, aerosol and cloud layers

were considered to be separated (Fig. 2(b)). Other conditions were not taken into consideration in this study. The aerosol and cloud types were defined using the Feature_Classification_Flags (FCF) derived from CALIPSO. Moreover, AOD and COD datasets from CALIPSO, and CF, CER and LWC datasets from CloudSat were used to explore the interaction between aerosol and cloud layers at different heights. Although the detection times of the two satellites have a gap of 12.5 seconds, they can detect the same cloud layer at almost the same time.

RESULTS AND DISCUSSION

Spatial Distributions of AOD_{ship}

AOD_{ship} is influenced by shipping-originated aerosols, such as primary particles, OC, EC and other secondary pollutants. Fig. 3 shows the spatial distributions of AOD_{ship} in spring (MAM), summer (JJA), autumn (SON) and winter (DJF). Clearly, the mean AOD_{ship} over the study area was approximately 0.17, 0.20, 0.15 and 0.13 in four seasons, and accounted for 23%, 30%, 36% and 25% of total AOD, respectively. The maximum AOD_{ship} appeared in summer due to relatively higher aerosol loading and atmospheric RH, during which increased particles from shipping emissions and their hygroscopic growth can magnify aerosol extinction to solar radiation (Xin *et al.*, 2014).

Over remote sea areas, in spring and summer, AOD_{ship} was generally higher mainly due to high RH and effective sunlight, while in autumn, AOD_{ship} was lower due to the prevailing northeast wind usually bringing clean air. Shen *et al.* (2019) also found low AOD during autumn over Yellow and Bohai Sea. In winter, the atmospheric circulation over the NECS was dominated by strong near-zone continental outflow in the middle atmosphere, and northwesterly winds prevailing in the boundary layer. This air current situation provides an effective approach to transporting pollutants originating from large discharge areas to the NECS after a long distance.

Table 2. The statistical results of AOD affected by ships and sea salt.

Season	Days
Spring (MAM)	37
Summer (JJA)	48
Autumn (SON)	63
Winter (DJF)	36

Fig. 2. Vertical features of aerosol and cloud layers from CALIPSO in cases of (a) 16 January and (b) 15 June.

Fig. 3. The spatial distributions of shipping-originated aerosol AOD_{ship} in spring (MAM), summer (JJA), autumn (SON) and winter (DJF) of 2014.

Relationships between Cloud Properties and AOD_{ship}

The effect of AOD_{ship} on cloud properties and the relationships between them without distinguishing aerosol-cloud overlap were discussed using MODIS 08 datasets. Fig. 4 shows the spatial and seasonal distributions of correlation coefficients of AOD_{ship} versus COT (top), CER

(middle; LWP is held fixed) and CF (bottom), where the black dot areas pass the significance test ($\alpha = 0.05$).

COT vs. AOD_{ship}

In general, COT decreased with increasing AOD_{ship} during all four seasons, which implies that clouds possibly

Fig. 4. The spatial distributions of correlation coefficients between AOD_{ship} and COT (top), CER (middle) and CF (bottom) in the spring, summer, autumn and winter of 2014. The black dot areas passed the significance test ($\alpha = 0.05$).

become optically thinner as more shipping-originated particles are carried into clouds by airflow (Fig. 4). Clearly, an obvious correlation between AOD_{ship} and COT existed over the offshore areas of East China. In spring and winter, a negative correlation existed between AOD_{ship} and COT over the southern areas of NECS, with correlation coefficients (R) of -0.42 and 0.51 , respectively. In the autumn, although AOD_{ship} over remote sea areas was low, the correlation between AOD_{ship} and COT over these areas was better than in the other three seasons.

Two possible effects are considered to be responsible for the above negative relationships. Firstly, cloud droplets evaporate due to the absorbing aerosols (such as black carbon) and then clouds become thinner, which is called the radiative effect (Liu *et al.*, 2017). Black carbon is a highly condensed carbonaceous residue arising from incomplete combustion and is a major light-absorbing component in atmospheric aerosols (Wu *et al.*, 2016). Secondly, the absorbing aerosols may affect the retrieved

COT from satellite due to the radiation effects and then decrease the cloud reflectance (Ten Hoeve *et al.*, 2011; Li *et al.*, 2014; Meyer *et al.*, 2015). Alam *et al.* (2014) also indicated that due to the effects of absorbing aerosols the detected cloud reflectance from the cloud top at visible wavelengths is usually smaller than expected. Meyer *et al.* (2013) pointed out that for the clouds in polluted marine boundary layer, adjusting the aerosol attenuation above the cloud can increase the average regional retrieved COT by approximately 18%. Thus, the radiative effect and the retrieval uncertainty play an important role in the relationship between COT and AOD .

CER vs. AOD_{ship}

There was a positive correlation ($R = 0.35$) between CER and AOD_{ship} in the winter but a weakly negative correlation ($R = -0.17$) in the summer over the NECS and a positive correlation in the spring and summer over the YS (Fig. 4). Feingold *et al.* (1999) found that slightly

soluble organic particles (SSO) and giant cloud condensation nuclei (CCN) present in the ship-originated aerosols, and even low concentrations of larger CCN emitted from ship can affect cloud properties. Yuan *et al.* (2008) concluded that there was a positive correlation between AOD and CER over East China due to the effects of SSO and larger CCN. Yuan *et al.* (2008) explained that the presence of SSO and larger CCN increased the AOD and decreased the total cloud droplets and thus CER increased according to the simulation of the 2-D Goddard Cumulus Ensemble model. The reason is that almost all SSO are difficult to be activated and hydrophobic as well as larger CCN prefer to absorb water vapor because of their lower critical supersaturation, leading to the deactivation of small aerosol particles (Rosenfeld *et al.*, 2002). Tang *et al.* (2014) observed that when AOD was greater than 0.3, positive correlations contrary to the Twomey effect between CER and AOD existed over the YS area in summer, whereas negative correlations consistent with the Twomey effect existed over the NECS area when AOD was less than 0.3. The present study showed that negative correlations between CER and AOD_{ship} appeared over the NECS for AOD_{ship} less than 0.2 during summer, whereas positive correlations between them over the YS for AOD_{ship} greater than 0.2.

CF vs. AOD_{ship}

There was a negative correlation ($R = -0.23$) between CF and AOD_{ship} during all four seasons over the coastal areas of the NECS due to more primary particles and a weakly positive correlation during the spring over the YS

due to aged particles (Fig. 4). The evaporation of cloud droplets caused by absorbing aerosols always makes clouds thinner (Liu *et al.*, 2017). For the positive correlation, the possible interpretation is that the retrieved AOD becomes higher with increased clear-sky reflectance close to cloud edges due to the 3-D effects (Várnai and Marshak, 2009). Other possible interpretations are higher humidity leading to greater aerosol volume near clouds (Twohy *et al.*, 2009) as well as convergence resulting in thicker aerosol layers (Mauger and Norris, 2007).

Differences between Separated and Mixed Aerosol-cloud Layers

In the view of climatology, in spite of correlation existing between cloud properties and AOD, the aerosol effects on cloud physical properties cannot be fully understood. Aerosols are often pinched into the cloud body to affect the microphysical properties of cloud droplets and participate in cloud formation and disappearance processes. Aerosol-cloud overlap is a typical situation used to investigate the direct influence of aerosol particles on clouds in the vertical direction through convection or entrainment. We examined the response of cloud physical properties to increasing AOD_{ship} for separated and mixed overlaps of aerosol and cloud layers based on CALIPSO and CloudSat datasets. Fig. 5 shows the relationship between major cloud parameters and AOD_{ship} under two aerosol-cloud overlap conditions. Commonly, in the case of aerosol-cloud mixed overlap, the cloud properties of COD, CER (LWC is held fixed) and CF had negative correlations with AOD_{ship}

Fig. 5. Scatter plots of cloud parameters versus AOD_{ship} for cases of vertical separated (red) and well-mixed (blue) aerosol-cloud layers.

(absolute $R > 0.3$). Whereas in the case of aerosol-cloud separated overlap, the above parameters showed weaker negative or positive correlations.

The relationship between COD and AOD_{ship} indicates that at mixed overlap, the evaporation of cloud droplets causes clouds to be thinner by shipping-originated absorbing aerosols. The relationship between CER and AOD_{ship} indicates that when uplifted shipping-emitted aerosols enter into the cloud body, the cloud droplets increase and thus CER decreases. However, when aerosol and cloud layers are separated, CER varies much less with AOD_{ship} , as there are no interactions between aerosols and clouds resulting from no aerosols directly participating in cloud microphysical processes. Liu *et al.* (2017) revealed that CF is smaller due to the presence of smoke. Shipping-originated aerosols are composed of completely smoke-containing fine absorbing particles and result in an increase of small droplets through the evaporation of cloud droplet water and the reduction of cloud cover due to the promotion of aerosol efficiency to sunlight absorption.

Meteorological Effects on AOD_{ship} and Cloud Properties

Feingold *et al.* (2001) pointed that aerosol hygroscopicity and PVV played an important role on aerosols' indirect effect. Wang *et al.* (2014b) demonstrated that the dynamic and thermodynamic processes in the cloud systems can influence the aerosol-cloud interactions. To investigate whether the aerosol (e.g., CCN) variations are relevant with systematic meteorological situations, we focused on the regional-scale changes of air vertical motion (PVV)

and RH. Based on the findings of the previous section, we will focus on the relationship between AOD_{ship} and cloud properties under well-mixed overlap conditions.

Relative Humidity

Relative humidity is an important factor that influences aerosol particle size and cloud development. High RH at the bottom of the cloud can influence the relationship between aerosols and clouds (Small *et al.*, 2011). According to RH at 950 hPa, we took $RH = 80\%$ as the cut-off value and analyzed the relationships of cloud properties versus AOD_{ship} at classified RH levels. Fig. 6 shows the relationships between cloud properties and AOD_{ship} for cases of low RH ($< 80\%$) and high RH ($> 80\%$).

As shown in Fig. 6, the correlation coefficients between cloud parameters and AOD_{ship} were generally higher at high RH (absolute $R > 0.3$) than at low RH. Meanwhile, the values of cloud parameters were higher at high RH than at low RH for the same level of AOD_{ship} . Obviously, there was a turning point of AOD_{ship} ranging from 0.3 to 0.4 for the correlation between cloud parameters and AOD_{ship} . The cloud parameters of COD, CER (LWC is held fixed) and CF decreased with increasing AOD_{ship} at $RH > 80\%$, but in contrast, at $RH < 80\%$, the results indicated that the ship-originated aerosols that were rolled into the cloud body could influence cloud properties by increasing the cloud condensation nuclei (CCN) number at low aerosol loadings and possibly by enhancing the warming of absorbing particles at high aerosol loadings. This implies that it is easy to form optically thicker and higher clouds

Fig. 6. Scatter plots of cloud parameters versus AOD_{ship} for cases of high RH (red) and low RH (blue) at 950 hPa.

under high RH condition. The possibility of cloud droplet activation as well as the growth of cloud droplets increases with RH (Jones *et al.*, 2009). This indicates that high RH contributes to larger cloud droplets because of higher water vapor content existing in the atmosphere.

Pressure Vertical Velocity

PVV at 750 hPa is commonly used as a meteorological indicator of atmospheric dynamic state. For low index cloud regimes, PVV is treated as a good proxy of the mean convective air mass flux simulated by Fan *et al.* (2011). The presence of upward motion is favorable for cloud formation due to the ambient environment, which is called negative PVV, and can also promote aerosol and cloud interactions (Jones *et al.*, 2009). In the present study, high frequency of mixed aerosol-cloud mixture occurred at a low and medium level of cloud height less than 6 km. Thus, PVV at 750 hPa was chosen to represent atmospheric dynamic state. Fig. 7 shows the relationships between cloud properties with AOD_{ship} for cases of negative PVV (< 0) and positive PVV (> 0).

As shown in Fig. 7, the correlation coefficients between cloud parameters and AOD_{ship} were generally higher at negative PVV than at positive PVV. Meanwhile, the values of cloud parameters were higher at negative PVV than at positive PVV for the same level of AOD_{ship} . This implies that negative PVV favors the formation of optically thicker clouds as well as larger cloud droplets. Additionally, negative PVV can also promote cloud development and

increase cloud cover.

CONCLUSIONS

The effects of shipping emissions on the micro- and macrophysical properties of clouds in marine areas near East China were investigated from a climatological perspective using multi-satellite datasets for the year 2014. Statistical analysis showed that although AOD_{ship} , seasonally averaged, was higher in spring and summer, its contribution to the total AOD was higher in summer and autumn. The quantity of shipping-originated aerosols (based on AOD_{ship}) is correlated with the major micro- and macro-scale physical cloud properties (COT, CF and CER), in spite of differences in time and place (e.g., offshore or remote sea areas). Irrespective of the thermodynamic structures, such as water vapor and upward air motion, a low cloud bottom facilitates the updraft of shipping-originated aerosols and contributes to the formation and development of clouds above the aerosol layer. Overall, shipping emissions may affect clouds by increasing the number of cloud condensation nuclei (CCN) at low aerosol loadings and by warming absorbing particles at high aerosol loadings.

How aerosols directly affect clouds and precipitation and thus the global climate remains a challenging question due to the complex mechanisms driving atmospheric thermodynamic conditions and aerosol-cloud interaction. In the future, we will extend our research on shipping-originated aerosol effects in marine areas by applying numerical

Fig. 7. Scatter plots of the cloud parameters versus AOD_{ship} for cases of negative PVV (red) and positive PVV (blue) at 750 hPa.

models and big data technology (Wang and Zhao, 2016; Wu, 2016), which supply relatively complete descriptions of physical and chemical processes in the atmosphere, and by assessing the influence of the major ship-emitted chemical species.

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