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Positive relationship between liquid cloud droplet effective radius and aerosol optical depth over Eastern China from satellite data

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HIGHLIGHTS

- We studied the first aerosol indirect effect over Eastern China.
- Twomey effect was observed over ocean.
- Positive correlations between aerosol optical depth and cloud effective radius were found over land for most seasons.
- We attributed the positive effect to the associated changes in relative humidity and wind fields.

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ABSTRACT

Correlations between water cloud effective radius (CER) and aerosol optical depth (AOD) from the Moderate Resolution Imaging Spectroradiometer (MODIS) are examined over seven sub-regions in Eastern China for 2003–2012. Water phase cloud is defined as having a cloud top pressure greater than 800 hPa. Significant negative correlation coefficients ($r = -0.79 \sim -0.94$) between AOD and CER are derived over the East Sea and the South China Sea for grid cells with AOD < 0.3. However, positive correlations ($r = 0.01$–0.91) are calculated for cells with AOD > 0.3. In contrast, significant positive correlations ($r = 0.67$–0.95) are derived over the Eastern China mainland and Yellow Sea. Further analysis for North China Plain shows that variations in wind speed and relative humidity may account for such positive correlations. Southerly winds carry high levels of pollutants and abundant water vapor, resulting in coincident increases in both AOD and CER in North China Plain, while the northerly winds transport dry and clean air from high latitudes, leading to decreases in AOD and CER. Both processes contribute to the positive correlations between AOD and CER over Eastern China, suggesting that the influence of background weather conditions need to be considered when studying the interactions between aerosol and cloud.

1. Introduction

By acting as cloud condensation nuclei (CCN), aerosol particles increase cloud droplet number concentration (Ramanathan et al., 2001; Andreae, 2009), limit cloud droplet size at constant liquid water path, and increase cloud albedo; termed the first aerosol indirect effect (FAIE) or Twomey effect (Twomey, 1977; Breon et al., 2002; Feingold et al., 2003). Satellite measurements can provide global observations for statistical studies of the relationships between aerosol and cloud properties. The slopes of correlation analysis evaluated for some parameters have shown some skill in representing the magnitude of FAIE (Nakajima et al., 2001; Feingold et al., 2001; Breon et al., 2002; Sekiguchi et al., 2003; Kaufman et al., 2005; Huang et al., 2006). Uncertainties in the characterization of FAIE include measurement biases and factors such as the size distribution, chemical composition, and mixing state of aerosol

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Table 1
Previous studies of the first aerosol indirect effect (FAIE) based on satellite data. IE (unitless) refers to the indirect effect and is the slope of the cloud effective radius (CER) to aerosol index (AI) or to aerosol optical depth (AOD). RF is the estimated radiative forcing due to IE at the top of atmosphere. *Na* is particle number, and *Nc* is cloud droplet number.

Reference	Instrument	Parameter	Region	Result
Nakajima et al., 2001	AVHRR	CER, AI	Global ocean	RF = -0.7 to -1.7 W m^{-2}, IE = 0.17
Feingold et al., 2001	AVHRR	CER, AOD	Amazon	IE = 0.12–0.38
Breon et al., 2002	POLDER	CER, AOD, AI	Global ocean	IE = -0.085 W m^{-2}
Sekiguchi et al., 2003	AVHRR POLDER	CER, *Na*	Global	RF = -0.6 to -1.2 W m^{-2}
Kaufman et al., 2005	MODIS	CER, AOD	Atlantic	ΔCER = 10%–30%
Yuan et al., 2008	MODIS	CER, AOD	China, Mexico	IE = -2.73–2.03
Costantino and Breon, 2013	MODIS CALIPSO	Vertical profiles	Angola coast	IE = -0.03 to -0.15
Lohmann and Lesins, 2002	POLDER	CER, AI	Global	RF = -0.85 W m^{-2}
Quaas et al., 2006	MODIS POLDER	CER, AOD, AI	Global	RF = -0.3 to -0.5 W m^{-2}
Penner et al., 2012	MODIS CERES	*Nc*, AI TOA fluxes	North Pacific Ocean	RF = -1.8 to -2.2 W m^{-2}

particles, as well as meteorological conditions (Lohmann and Feichter, 2005; Mauger and Norris, 2007). To date, few studies of FAIE using satellite data have been performed over China, where air pollution is heavy and the meteorological environment is complex due to the Asian monsoon (Sekiguchi et al., 2003; Yuan et al., 2008). In this work, FAIE is investigated, and the influence of meteorological conditions is analyzed over seven sub-sectors of Eastern China and the surrounding waters using aerosol and cloud properties observed by the Moderate Resolution Imaging Spectroradiometer (MODIS) and meteorological variables from the European Centre for Medium-Range Weather Forecasts (ECMWF) reanalysis.

An array of approaches has been used to retrieve aerosol and cloud properties from observations. For example, aerosol number concentrations can be measured directly by instruments onboard aircraft (Warner and Twomey, 1967; Andreae et al., 2004) or estimated using aerosol scattering coefficients measured from the ground (Feingold et al., 2003). They may also be represented by column integrated aerosol optical depth (AOD) or aerosol index (AI) measured by satellites (Nakajima et al., 2001; Breon et al., 2002; Kaufman et al., 2005; Huang et al., 2006). Compared with the former two approaches, satellite observations provide a large sample size with high coverage in both space and time, and as a result are more applicable to estimate FAIE on global or regional scales, as outlined in Table 1. In these studies, negative correlations between AOD and cloud effective radius (CER) have been found over oceans (Nakajima et al., 2001; Breon et al., 2002; Kaufman et al., 2005), and positive correlations have been observed over China, the Gulf of Mexico and the Mediterranean Sea (Sekiguchi et al., 2003; Myhre et al., 2007; Yuan et al., 2008). The relationships of aerosol and cloud derived from satellite measurements can be used to constrain climate model simulation and decrease uncertainty of FAIE by a factor of two (Lohmann and Lesins, 2002; Quaas et al., 2006; Penner et al., 2012).

Quantifying FAIE is very uncertain because it is associated with both physical and chemical properties as well as the mixing state of aerosol particles (Nenes et al., 2002; Chen and Penner, 2005; Dusek et al., 2006; Wang et al., 2008), vertical distributions of aerosol and cloud (Costantino and Breon, 2013), and meteorological conditions like relative humidity (RH), wind fields, updraft velocity and lower troposphere static stability (Loeb and Schuster, 2008; Kaufman et al., 2005; Mauger and Norris, 2007; Koren et al., 2010; Wang et al., 2010). For example, large-scale low-level convergence increases cloudiness as well as aerosol number concentration, and relative humidity enhances cloud droplet growth and deliquescence (Mauger and Norris, 2007; Koren et al., 2010). Such effects of meteorology are important, but difficult to isolate from broad observations, like those from satellite.

Pollution in China is characterized by a wide variety of chemical compositions and is influenced by the Asian monsoon, especially in the eastern part of the country (Ding and Chan, 2005; Hu and Ding, 2010; Zhang et al., 2012). Sekiguchi et al. (2003) demonstrated a positive correlation between CER and AI from Polarization and Directionality of the Earth's Reflectances (POLDER) measurements in a $17.5^\circ \times 17.5^\circ$ grid in China. Yuan et al. (2008) confirmed this phenomenon using AOD and CER from MODIS. Using a model simulation, they attributed the positive FAIE over the Gulf region to the influence of slightly soluble organic aerosols and sea salt aerosols, which accounts for higher AOD but delays the activation of small aerosol particles that results in higher CCN and lower CER. However, more detailed analysis is required to explain positive FAIE in China, because previous studies covered a large domain characterized by complex aerosol compositions and varying meteorological conditions, which together may affect the seasonality of cloud systems, aerosol hygroscopic growth, and their correlations.

In this study, FAIE over Eastern China and surrounding waters are examined using aerosol and cloud retrievals from MODIS and meteorological parameters from reanalysis data and in situ measurements. Eastern China is divided into seven sub-sectors with relatively common aerosol properties and meteorological conditions. Coefficients and slopes of the correlation between AOD and low-level liquid cloud effective radius with cloud-top pressure (CTP) greater than 800 hPa are calculated for each region, and the influences of winds and RH on correlations between AOD and CER are analyzed. Datasets used in this study are introduced in Section 2. Results of the study and effects of atmospheric circulations are described in Sections 3 and 4, respectively, and the conclusion and discussion are presented in Section 5.

2. Data and methodology

Daily AOD, CER, and CTP observed by MODIS onboard the Aqua satellite from 2003 to 2012 on $1^\circ \times 1^\circ$ grids are used in this work. For the retrieval of aerosol and cloud properties, MODIS identifies the state of each pixel (clear or cloudy) based on spatial variability (Ackerman et al., 1998). For clear pixels, two independent algorithms are applied to retrieve aerosol properties: the Deep Blue method over land using reflectances at 0.47, 0.66, and 2.13 µm and the dark target method over ocean using seven bands ranging from 0.47 to 2.13 µm (Remer et al., 2005). MODIS Level 2 AOD has a footprint of 10 × 10 km and an overall uncertainty of $\pm 0.05 \pm 0.15\,\tau$ over land and $\pm 0.03 \pm 0.05\,\tau$ over ocean (Remer et al., 2005). The Deep Blue algorithm decreases the influence of albedo effects over bright surfaces, thus has high accuracy over land, except over dust and snow surfaces. Thus, our target region does not include Western China, which is desert in Xinjiang and frequently snow covered on the Tibetan Plateau.

By comparison, MODIS retrieves cloud physical and optical properties using 14 spectral bands. Cloud phase (water or ice) is determined based on the distinct differences in absorption at 1.64 µm between water and ice (King et al., 2003). CER is retrieved at 1 × 1 km resolution using absorption at 2.13 µm in combination with information from non-absorbing bands at 0.65, 0.86, and 1.24 µm over land, ocean, and ice surfaces (Platnick et al., 2003).