Opposite Aerosol Index-Cloud Droplet Effective Radius Correlations Over Major Industrial Regions and Their Adjacent Oceans

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Abstract The Moderate Resolution Imaging Spectroradiometer (MODIS) C6 L3 and the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim reanalysis data from 2003 to 2016 are employed to study aerosol-cloud correlations over three industrial regions and their adjacent oceans, as well as explore the impact of meteorological conditions on the correlations. The analysis focusing on liquid and single-layer clouds indicates an opposite aerosol-cloud correlation between land and ocean; namely, cloud effective radius is positively correlated with aerosol index over industrial regions (positive slopes), but negatively correlated over their adjacent oceans (negative slopes), for a quasi-constant liquid water path. The positive slopes are relatively large under low lower-tropospheric stability (LTS; weakly stable condition), but much weaker or even become negative under high LTS (stable conditions) and high liquid water path. The occurrence frequency of cloud top height (CTH) and LTS suggests that positive correlations are more likely corresponding to relatively high CTH and low LTS, while negative to low CTH and high LTS.

Plain Language Summary Aerosol-cloud interactions play an important role in climate prediction, but remain to have large uncertainties. The major industrial regions generally exhibit relatively high aerosol concentrations, and how aerosols impact cloud properties, and how aerosol-cloud interactions modulate cloud and precipitation processes, is an interesting and challenging topic. In this study, a new retrieval from long-term satellite data is employed to examine the correlation between aerosol concentration and cloud droplet size over the three industrial regions and their adjacent oceans. The results show the opposite aerosol-cloud correlation between land and ocean. Positive slopes are more likely associated with high cloud top height and low lower-tropospheric stability. This finding is very useful to the communities for an accurate prediction of weather and climate.

1. Introduction

Aerosol particle concentrations have increased dramatically since preindustrial due to anthropogenic emissions, particularly over northern industrial regions. They influence climate change by directly scattering and absorbing incoming solar radiation and indirectly acting as cloud condensation nuclei (CCN), altering cloud properties, and precipitation (Albrecht, 1989; Twomey, 1977). According to Twomey (1977), the increase of aerosol particles results in more and, for a constant cloud liquid water path (LWP), smaller cloud droplets, that is, cloud effective radius (CER) decreases with increasing of aerosol concentrations. Many previous studies have confirmed this negative relationship between aerosol number and CER, either from aircraft observations (Kleinman et al., 2012; Pawlowska & Brenguier, 2000; Roberts et al., 2008; Werner et al., 2014; Wilcox et al., 2006) or satellite data (Bréon et al., 2002; Chen et al., 2014; Christensen et al., 2016; Costantino & Bréon, 2010, 2013; Kaufman et al., 2005; Koren et al., 2005, 2010; Lihavainen et al., 2010; Nakajima et al., 2001; Quaas et al., 2008; Zhu et al., 2015).

In recent years, however, quite a few studies found that this relation is not always true; instead, the negative correlation between aerosol particles and CER is not found distinctly or could be even positive; that is, CER increases with aerosol number (Bulgin et al., 2008; Grandey & Stier, 2010; Liu et al., 2017; Sekiguchi et al., 2003; Tang et al., 2014; Wang et al., 2014, 2015; Yuan et al., 2008). It is acknowledged that aerosol-cloud interactions are quite complex processes, influenced by both aerosol microphysics (size and chemical
composition) and meteorological conditions. The studies by Dusek et al. (2006) and Zhang et al. (2011) found that particle size is dominant to determine aerosol activation, but many studies demonstrated that the chemical composition is also critical (Almeida et al., 2014; Ervens et al., 2005; Lance et al., 2004; McFiggans et al., 2006; Nenes et al., 2002; Wang et al., 2008). In addition, dynamical or meteorological conditions will strongly affect aerosol-cloud interactions by changing vertical velocity or wind shear (Gryspeerdt et al., 2014; Fan et al., 2009; Koren et al., 2010; Loeb & Schuster, 2008; Mauger & Norris, 2007; Saponaro et al., 2017; Su et al., 2010; Tang et al., 2014; Wang et al., 2014). Overall, negative correlations are normally found over oceans while positive over land. Yuan et al. (2008) explored the positive correlation using Moderate Resolution Imaging Spectroradiometer (MODIS) satellite products and speculated that a positive correlation is possibly due to slightly soluble organics particles, which induced a decrease of aerosol activation. Wang et al. (2014) investigated aerosol-cloud interaction over Eastern China by using both MODIS satellite and National Centers for Environmental Prediction reanalysis data and concluded that negative correlations are usually found when atmospheric stability is relatively high while positive correlations are more likely to be found when the atmosphere is relatively unstable. Some recent studies found that the correlation is probably also related to cloud type; for example, Chen et al. (2016) employed CloudSat satellite and ERA-Interim reanalysis data to identify the impact of aerosols on the vertical development of clouds and found out that aerosols inhibit the cloud development for shallow cumulus clouds, while enhance deep cumulus, nimbostratus, and deep convective clouds. Gryspeerdt and Stier (2012) found that the strongest positive correlation occurs in the stratiform cloud regimes, while the negative one is analyzed for the shallow cumulus cloud regimes. For the northern industrial regions, anthropogenic emissions are much higher than over other continental regions; thus, how aerosols impact cloud formation and development and further climate change over such regions is a vital question to address, but substantial uncertainties still remain.

In this study, we chose three regions with strong anthropogenic emissions in the Northern Hemisphere, namely, East China (EC), East U.S. (EU), and West Europe (WE), and their adjacent oceans (ECO, EUO, and WEO), to systematically explore the aerosol-cloud correlations over land with strong anthropogenic emissions and over relatively clean ocean by using both MODIS satellite data and ERA-Interim reanalysis data. The paper is organized as follows: the data descriptions of both satellite and meteorology and data processing are presented in section 2; the aerosol-cloud correlations over the study regions are discussed in section 3. Possible explanations and discussions are summarized in section 4.

2. Data

The data used in this study include the MODIS Collection 6 Level 3 (C6 L3) product, which provides aerosol (Levy et al., 2013) and cloud properties (Platnick et al., 2017) at 1° × 1° spatial resolution and daily time resolution, as well as European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-Interim reanalysis data, which provides meteorological fields at spatial resolution from 0.125° to 2.5° and at 4 times (00, 06, 12, and 18 UTC) every day (Dee et al., 2011). The total 14 years (2003–2016) of these data are used for analysis.

2.1. MODIS

Although it is essential to have coincidental aerosol and cloud properties in order to examine their correlations, many satellite retrievals do not provide such data since aerosol retrievals are usually conducted when cloudy conditions are excluded. However, studies such as Bréon et al. (2002), Anderson et al. (2003), and Avey et al. (2007) showed that in the case of daily products at 1° × 1° resolution, it is possibly unnecessary to individually couple the aerosol and cloud measurements. Therefore, aerosol and cloud data retrieved from cloudy and clear, respectively, pixels within a 1° × 1° in this study are assumed to be co-located. The MODIS C6 L3 product includes cloud microphysical parameters (CER, LWP, and cloud optical depth) with statistics (mean, minimum, maximum, and standard deviation) determined at three different wavelengths (1.6, 2.1, and 3.7 μm) for each cloud phase (liquid, ice, and undetermined) separately. We filtered the MODIS cloud data according to the criteria employed by Saponaro et al. (2017) to make sure the data used are only for liquid phase and single layer clouds, and also for nonprecipitating cases. The daily AOD product and cloud parameters from January 2003 to December 2016 were employed for the statistics in order to obtain statistically significant results from the samples large enough.
Figure 1. The computed slopes of CER versus Al on log-log scale, in which both CER and AI are stratified according to LWP.

Table 1

| Region | LWP (g m⁻²) | 20–60 | 60–100 | 100–140 | 140–180 | 180–220 |
|--------|-------------|-------|--------|---------|---------|---------|
| ECO    | R²         | t     | t      | t       | t       | t       |
|        | 0.32       | 0.33  | 0.36   | 0.43    | 0.62    | 0.62    |
| EUO    | R²         | t     | t      | t       | t       | t       |
|        | 0.01       | 0.55  | 0.58   | 0.49    | 0.75    | 0.75    |
| WEO    | R²         | t     | t      | t       | t       | t       |
|        | 0.16       | 0.76  | 0.89   | 0.89    | 0.90    | 0.90    |
| EC     | R²         | t     | t      | t       | t       | t       |
|        | 0.74       | 0.62  | 0.41   | 0.33    | 0.41    | 0.41    |
| EU     | R²         | t     | t      | t       | t       | t       |
|        | 0.91       | 0.90  | 0.90   | 0.40    | 0.12    | 0.12    |
| WE     | R²         | t     | t      | t       | t       | t       |
|        | 0.85       | 0.83  | 0.54   | 0.04    | 0.13    | 0.13    |

Note. The coefficients of determination (R²) is listed, and the cases at a statistically significant level (according to a Student’s t test, α = 0.05) are represented in circles, otherwise in crosses.

2.2. Meteorology

To assess the effect of meteorological conditions on cloud properties, the ECMWF ERA-Interim reanalysis data were applied to derive the lower-tropospheric stability (LTS). The LTS is computed as the difference between the potential temperature at 700 hPa and at the surface (Klein & Hartmann, 1993), describing the magnitude of the inversion strength for the lower troposphere.

3. Aerosol-Cloud Correlations

3.1. Aerosol Index and Cloud Properties

Aerosol index (Al), in comparison to aerosol optical depth (AOD), is believed to be a better parameter to represent aerosol when examining its correlation with cloud, because it includes the information of aerosol size, which is critical for cloud formation and properties (Penner et al., 2011; Stier, 2016). The Al is used as a proxy for the fine mode aerosol particles, which have a larger contribution to the CCN than the coarse-mode particles (Nakajima et al., 2001). The Al is derived based on AOD and Ångström exponent; the former is provided by the MODIS C6 product, and the latter is calculated from AOD at wavelength 460 and 660 nm.

According to Twomey, more but, for constant liquid water content, smaller cloud droplets form as aerosol particle concentrations increase. Both Al and CER here are therefore stratified according to LWP in order to examine the correlations between CER and Al at smaller range of LWP (quasi-constant LWP). CER varies with Al over land, and ocean at various cloud top layer (650–750 hPa, 750–850 hPa, 850–950 hPa, and 650–950 hPa; Figure S1) shows that CER decreases with increasing Al over ocean, but CER increases with Al over land. The positive correlations can be found at various vertical levels, which is most distinct at smaller LWP and weaken with the increase of LWP. The computed slopes from linear regressions of CER versus Al on log-log scale with LWP-stratified for the layer 650–950 hPa over three anthropogenic regions and their adjacent oceans are summarized in Figure 1, which clearly show that for a given LWP, slopes are normally negative over ocean but positive over land. The opposite correlation over land has been reported, though not explained, in a few previous studies (Grandey & Stier, 2010; Liu et al., 2017; Tang et al., 2014; Wang et al., 2014, 2015; Yuan et al., 2008). It is interesting to see that as LWP increases, the positive slopes over land become weaker while negative slopes over ocean change slightly. The associated statistics listed in Table 1 demonstrated that the slopes of the regression over land are in all cases—but four (both for 140–180 and 180–220 g/m²)—positive at a statistically significant level (according to a Student’s t test, α = 0.05). It is argued that the MODIS retrieval of Ångström exponent over land is problematic (Sayer et al., 2013); thus, a similar analysis as discussed above but using AOD as aerosol proxy are conducted, which is shown in Figure S2 and Table S1. It is clearly demonstrated that the statistical results, particularly the signs and magnitudes of slopes, though change slightly, do not influence our conclusions.

The cloud properties as a function of cloud top pressure (CTP) are presented in Figure 2, in which all cloud data are grouped into five classes according to Al. The groups are classified to make sure that each group has roughly same number of the samples. Koren et al. (2005) previously suggested an enhancement of cloud vertical structure over the North Atlantic due to aerosol loading. Saponaro et al. (2017) also found that the lowest CTP corresponds to the highest classes of Al over the Baltic Sea region. This is not evident over three anthropogenic regions but seems true over the adjacent oceans (Figure 2), especially ECO and EUO, where the lowest CTP values (highest cloud top height, CTH) correspond to highest classes of Al. The highest cloud fraction (CF) is overall consistent with the highest classes of
Figure 2. (top row) Cloud fraction (CF) and (bottom row) liquid water path (LWP) as a function of cloud top pressure (CTP) over the three regions. Cloud properties are grouped into five classes according to AI.

Figure 3. (a) The computed slopes of CER versus AI on log-log scale at (left) low, (middle) medium, and (right) high LTS conditions, where all data are stratified by LWP. The white box represents NA; (b) CTP varies with CF and LWP under low, medium, and high LTS conditions.
AI, possibly indicating an enhancement of cloud formation due to aerosols, which is evident over all three adjacent oceans. An alternative explanation is covariation of CF and AI, both responding to relative humidity variability (Gryspeerdt et al., 2016; Quaas et al., 2010). Over land, however, although CF increases with AI over three continents, it is more evident over EC. LWP exhibits the opposite relationship with AI, that is, LWP increases with AI over WE while it decreases over EC. Over ocean, LWP, in general, is smaller when AI increases, which is more evident over ECO and EUO, possibly because large cloud droplets more likely become to rain droplet and thus reduce the LWP. The LWP remains rather constant over WEO, where both CER and LWP are much smaller than in the other two regions.

3.2. Impacts of Thermodynamic and Dynamic Parameters on the Correlations

Thermodynamic and dynamic conditions are important on vertical transport of aerosol, activation process, and aerosol-cloud interaction. LTS is often used to represent the large-scale thermodynamic conditions. We computed LTS from the difference of potential temperature between 700 hPa and sea level according to the definition by Klein and Hartmann (1993). The slopes of CER versus AI on log-log scale under low, medium, and high LTS conditions, and stratified by different classes of LWPs, are shown in Figure 3a. The three LTS levels have the same number of samples (Table S2). It is obvious that positive slopes under low LTS (weak stable condition) are relatively large compared to medium (medium stable) and high LTS (stable condition). It is also noted that positive slopes could be much weaker or even become negative under high LTS and when LWP are relatively high. This may imply that when the lower atmosphere is very stable, an accumulation of aerosol particles induces a rapid increase of aerosol size and thus more likely to activate as CCN, in which water vapor is exhausted very quickly for both aerosol hygroscopic growth and cloud droplet formation. This may induce a large number of small cloud droplet and thus higher LWP. Figure 3b confirmed that more CF was found when the lower layer atmosphere is very stable (high LTS), which is discerned over three anthropogenic regions. Overall, the negative slopes over ocean are consistent at low, medium, and high LTS. We conducted the statistical analysis by stratifying the data according to three categories of vertical velocity at 850 hPa (ω), that is, updraft, weak vertical velocity, and downdraft (Table S3), to test the impact of dynamic conditions on the correlations, and the results do not show evident dependence of the correlations on ω (Figure S3).

It is known that aerosol emission and its microphysical properties such as chemical composition, size, and mixing state could have strong seasonal variability, which possibly influences the process of aerosol activation and thus CER versus AI correlations (Bhattu & Tripathi, 2015; Crosbie et al., 2015; McFiggans et al., 2006; Meng et al., 2014; Padró et al., 2012; Paramonov et al., 2013; Petters & Kreidenweis, 2007; Topping &
McFiggans, 2012; Q. Zhang et al., 2012; Zhang et al., 2016). Unfortunately, there are only station-based observational data available for aerosol microphysics, it is impossible currently to obtain aerosol chemical component, size, and mixing state on a regional scale. As aerosol microphysics exhibit significant seasonal variations, it is thus useful to look into the correlations in other seasons. In comparison to summer season, the slopes in winter exhibit some yet insignificant differences (figure omitted). The negative slopes over ocean are generally unchanged; however, the positive slopes found in summer over land are less pronounced in winter; in some cases, for example, while LWFs are relatively high, slopes could become negative.

4. Summary and Discussions

It should be pointed out that CER-AI (AOD) correlations can result from both real physical-chemical processes and artificial correlations in light of various data uncertainties. Although we apply very strict screening criterion in selecting the data to avoid this issue as much as possible (see section 2.1), it is still possible that the correlation is veiled by various artifacts. The potential artifact includes cloud contamination, cloud 3-D effect, aerosol humidity, and dynamic effect. Previous studies (e.g., Yuan et al., 2008) assessed the potential effect of partial cloud and concluded that cloud contamination is not likely the cause of the observed positive correlation between AOD and CER. It is found that 3-D effects tend to have stronger impact on CER at 2.1 μm (CER,2.1) than CER at 3.7 (CER,3.7; Zhang & Platnick, 2011; Z. Zhang et al., 2012). We apply CER,3.7, rather than CER,2.1, to conduct the similar analysis, and the results (Figure S4 and Table S4) show a slight difference, so it seems that cloud 3-D effect might not influence our conclusion significantly. Although quite a few previous studies have attempted to explore what factors contribute to such positive values as stated earlier, they often apply to some conditions but not working for others (Tang et al., 2014; Wang et al., 2014; Yuan et al., 2008).

As presented above, positive slopes occur over three industrial regions, with the highest values over EC and EU during the summer seasons, while positive correlations become less pronounced during winter season and over WE. As shown in Figure 2 the CTP over three continental regions are overall lower than 600 hPa, with the lowest of 500 hPa over EC, while CTP over the adjacent oceans are typically higher than 600 hPa, with the highest of 650 hPa over WE. The results in winter also show the significant variations of CTP. The occurrence frequency of CTH during the entire study period in summer and winter presents a significant difference between the continental and their adjacent oceans, and the distinct regional differences as well (Figure 4). In summer, the occurrence of low clouds over ocean is most frequent, especially over ECO and EUO, where 84% clouds occur at lower than 2 km. On contrast, more clouds occur with CTH higher than 2 km over the industrial regions. The smallest difference of CTH between land and ocean is found over WE. In winter, the differences of CTH between land and ocean are much smaller, but we can still found that the higher clouds over land occur more frequently than ocean.

As expected, the frequencies of the cloud under lower LTS (unstable) are higher over land than over ocean. Also, unstable clouds occur more frequently in summer than in winter, and the differences of frequencies between land and ocean are very small in winter. Therefore, it is suggested that positive correlations are more likely occur when clouds are relatively high and unstable, while negative correlations normally occur when clouds are relatively low and stable. We hypothesize that this might be associated with collision/coalescence and/or entrainment mixing processes. More studies on the physical mechanisms will be conducted in subsequent research.

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