Contrasting Aerosol Effects on Long-Wave Cloud Forcing in South East Asia and Amazon Simulated With Community Atmosphere Model Version 5.3

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Abstract Aerosols modify cloud microphysical and radiative properties and thus impact the shortwave and long-wave cloud forcing (LWCF). This study first reports the finding of contrasting aerosol effects on LWCF in South East Asia and Amazon in the Community Atmosphere Model version 5.3 (CAM5.3), which corresponds to the sum of LW indirect and semi-direct effects investigated in Ghan et al. (2012, http://doi.org/10.1175/jcli-d-11-00650.1). A series of numerical experiments are conducted to decompose the complex aerosol effects on LWCF. Our analysis indicates that the cooling (negative aerosol effects on LWCF) in Amazon is due mainly to the aerosol effects on warm clouds and the inhibition of vertical motion by the aerosol-induced radiative cooling. In contrast, the warming (positive aerosol effects on LWCF) in South East Asia is due mainly to the aerosol effect on homogeneous freezing, thus, reducing the ice particle size and prolonging the existence of ice cloud. Our results emphasize that a comprehensive analysis of integrated aerosol effects on both warm and ice clouds is necessary for better understanding the aerosol effects on LWCF.

Plain Language Summary Aerosol particles in the atmosphere modify the number and size of cloud droplets and ice particles, and impact the cloud top height and extension, which modulate the thermal energy absorbed and emitted by clouds. We carried out a series of global climate model experiments and found that an increasing number of aerosol particles result in opposite effects in two regions. A cooling effect in Amazon is due to the enhanced number and reduced size of water cloud droplets. The shielding effect of water cloud cools the surface, thus, makes the cloud hardly develop higher. On the other hand, the warming effect in South East Asia is due mainly to the reduced ice particle size, prolonging the existence of the ice cloud. Our results indicate that the integrated aerosol effects on both water and ice clouds are complex, yet significant in understanding their impacts on cloud and energy in the atmosphere.

1. Introduction

Aerosol-cloud interactions (ACI) are among the least confident factors in climate models in projection of future climate change (Stocker, 2014, pp. 571–657). Aerosol particles affect the climate system directly by scattering, absorbing and emitting solar and thermal radiation, and indirectly by acting as cloud condensation nuclei (CCN) and ice nuclei (IN) to modify cloud microphysical and radiative properties. The semi-direct effect is a consequence of the direct effect of absorbing aerosols to modify cloud properties in response to thermodynamic changes due to aerosols (Bond et al., 2013), and regarded as a rapid adjustment to the Radiative Forcing from Aerosol-Radiation Interactions (RFari), also a part of Effective RFari (ERFari; Figure 7.3 in Chapter 7 of Intergovernmental Panel on Climate Change [IPCC], 2013). In the past several decades, cloud albedo enhancement (albedo effect, Twomey, 1991) and lifetime increase (lifetime effect, Albrecht, 1989) of warm stratus/stratiform clouds have received the most attention. Nevertheless, the uncertainty of both effects on warm clouds is still high (IPCC, 2013). According to the definition in IPCC (2013), the instantaneous effect on cloud albedo due to changing concentrations of CCN and IN is referred as the Radiative Forcing from aerosol-cloud interaction (RFaci). All subsequent changes to the cloud lifetime and thermodynamics are rapid adjustments, and contribute to the Effective RFaci (ERFaci). Compared to warm clouds, the aerosol effects on cirrus clouds (T < 35°C) are even more uncertain and controversial (Lohmann & Feichter, 2005; Seinfeld et al., 2016), although it is qualitatively known that cirrus clouds...
reduce the outgoing long-wave radiation at the top of the atmosphere (TOA; Ramanathan & Collins, 1991; Rossow & Schiffer, 1999) and result in a warming effect on the earth-atmosphere system. The increased anthropogenic aerosols from preindustrial (PI) times to present day (PD) may cause various changes in cloud properties (Ghan et al., 2012; Liu et al., 2009; Penner et al., 2009), and, thus, impact the ability of cirrus clouds in modulating the long-wave cloud forcing (LWCF) at the TOA.

One challenge in modeling cirrus in climate models is to represent the competition of the heterogeneous freezing and homogeneous freezing (Penner et al., 2009), which leads to the uncertainty in estimating the ice crystal number and size, and hence cloud lifetime and long-wave emissivity. For the source of ice nucleating particles (INPs), mineral dust and some biological particles exhibit a high degree of ice nucleation activity, but less clear at present is the extent to which kind of particles serve as INPs. The role of different aerosol types in nucleating ice at different threshold values of humidity is also uncertain (DeMott et al., 2011). In addition, updraft and moisture supply, including the water consumption by growing ice, can sometimes exert a stronger control on ice formation than the availability of INPs (Seinfeld et al., 2016).

Furthermore, aerosol effects on the radiative forcing of both warm and cirrus clouds are related to multiple mechanisms that involve interacting cloud microphysics, thermodynamics, and surface radiation flux, making it a challenge to find whether the aerosol effects on LWCF is positive or negative in current general circulation models (GCMs; Levy et al., 2013). Finding physical explanations is even more challenging. A wide range of the estimates of the aerosol short-wave and long-wave radiative forcing have reported in climate models (IPCC, 2013). For example, Ghan et al. (2012) estimates, by using CAM5.1 with the ice nucleation scheme of Liu and Penner (2005), that the resulted LW effects range from 0.27 to 0.54 W/m² by using different aerosol modules. Kooperman et al. (2012) obtains a LW effect of 0.35 W/m² with the same model as Ghan et al. (2012) but applying nudging technique or conducting the simulation with a longer period to reduce the possible noises caused by the model variability. Simulations of ECHAM-HAM2 (ECMWF Hamburg atmospheric general circulation model with Hamburg Aerosol Module version 2) model with the ice nucleation scheme from Karcher and Lohmann (2002a, 2002b) generate a LW effect of 0.67 ± 0.19 W/m² (Zhang et al., 2014). However, most studies about aerosol effects on LWCF reported the global mean estimates of the forcing, but did not show the spatial distribution or analyze the mechanism comprehensively.

Aerosol induced changes in cirrus cloud cover, cloud top temperature, and surface temperature in clear-sky can alter the upward long-wave radiation fluxes at TOA. In GCM, the uncertainty of the aerosol effects on LWCF is mainly investigated by using various kinds of aerosols to act as INPs in different parameterizations of ice nucleation (Gettelman et al., 2012; Penner et al., 2009; Shi & Liu, 2018) or by applying different cirrus clouds macrophysics scheme and modifying the microphysics scheme (Wang et al., 2014). For example, Liu and Shi (2018) analyzed the three commonly used ice nucleation parameterizations (Barahona & Nenes, 2008; Karcher & Lohmann, 2002a, 2002b; Liu & Penner, 2005, hereafter abbreviated as BN, KL, and LP schemes, respectively) by running a cloud parcel model and a GCM. Their results show that the uncertainty of aerosol effects on LWCF roots partly from the poor understanding of ice nucleation parameterizations. However, aerosols can influence the cloud properties not only by acting as CCN and IN but also by changing the stratification and stability of the atmosphere (rapid adjustments to RFaxi and RFaci). The entangled aerosol effects add complexity in analyzing the total aerosol radiative forcing (e.g., Huang et al., 2014). It is not clear what mechanism of ACI is responsible to the model simulated LWCF.

This work is conducted to address the LWCF challenges with CAM5.3 by decomposing the aerosol effects on LWCF and examining the spatial distribution of LWCF changed from PI to PD. Section 2 provides a brief description of model and setup of experiments. Section 3 presents the simulation results and analysis of different aerosol effects on LWCF and gives a short summary. Discussion on the limitation of the current study are given in Section 4.

2. A Model Description and Experimental Design

2.1. Model Description

CAM5.3 is a state-of-the-art global atmospheric model (Neale et al., 2012, p. 289) that has been widely applied to aerosol-climate studies. In this study, all the simulations are run with standalone CAM5.3 and driven repeatedly by climatological sea surface temperature (SST) and sea ice (SIC). SST and SIC data are
provided by Hadley Center as monthly means averaged over 1981–2001, which are interpolated to the model time step. Horizontal winds are nudged to 6-h ERA-Interim reanalysis of year 2000 (nudge technique is explained in the next subsection). All model simulations are integrated for 6 years, and the last 5 years results are analyzed in this study. The greenhouse gas concentrations are fixed. The surface concentration of carbon dioxide is 367 ppm (Neale et al., 2012). Model resolution is 0.9 × 1.25° horizontally and 31 layers vertically.

The input aerosol data in CAM5.3 follow the IPCC AR5 configurations, including the emissions of sulfate, primary organic carbon, black carbon (BC), sulfur dioxide (SO2), dimethyl sulfide, and volatile organic compounds. The emission datasets of aerosol and precursor gases are given for PI (year 1850 representing the PI) and PD (year 2000 representing the PD) scenarios, respectively. Sulfate, BC, and organic aerosols (sum of primary and secondary organic aerosols) increase substantially from PI to PD. For dust and sea salt aerosols, parameterized emission schemes are applied in CAM5.3 to online calculate the emitted amount, which are largely dependent on wind speed at the height of 10 m. Therefore, the relative changes in dust and sea salt burden from PI to PD are insignificant with the nudging wind field.

Aerosol mass concentration, number concentration, and size distribution are simulated online with the 7-mode Modal Aerosol Module (MAM7) in CAM5.3 (Liu et al., 2012). Different aerosol species are internally mixed in each mode, but externally mixed among different modes. Hygroscopicity of total aerosol in each mode is calculated by weighting the volume of each aerosol species. Aerosol optics in each mode is given in a precalculated look-up table, including aerosol extinction coefficient, single scattering albedo, and asymmetry factor in shortwave bands and aerosol absorption coefficient in long-wave bands. The look-up table provides aerosol optics data in prescribed particle size ranges, relative humidity (RH) bins, and for different aerosol species. Modeled aerosol optical properties are obtained by interpolating to the online calculated particle dry size parameter and RH values, to account for the humidity dependence of optical properties for soluble aerosols (sulfate, sea salt, hydrophilic BC and organic aerosol, and sulfate coated dust). Aerosol optical properties are accounted in RRTMG (the general circulation model version of the Rapid Radiative Transfer Model) for simulating shortwave radiation flux and heating rate at each layer, which will influence on the temperature and humidity profiles, atmospheric stability and cloud distribution in the next model time step.

In CAM5.3, soluble aerosols can act as CCN. Parameterization of activation process follows Abdul-Razzak and Ghan (2000), by accounting for the influences of size distribution, number concentration, and hygroscopicity of aerosols and subgrid vertical velocity in each model grid cell. Both increased soluble aerosols and strengthened vertical velocity will promote the aerosol activation and enhance the number of cloud droplets.

For ice nucleation, parameterization of homogenous freezing follows Liu and Penner (2005) and the nucleated ice particle number depends on the number concentration of pure sulfate aerosols (act as IN), atmospheric temperature, and vertical velocity. The parameterization of heterogeneous ice nucleation follows Liu et al. (2007), in which dust aerosol acts as IN. BC is also potential IN but not considered in the current CAM5.3, because the simulated cloud forcing is unrealistically high when including the nucleation of soot (Gettelman et al., 2012). Comparing to the homogeneous freezing, the heterogeneous nucleation requires lower supersaturation, and, thus, homogeneous freezing could be suppressed as a result of the competition of water vapor between homogeneous and heterogeneous nucleation. Lastly, no aerosol effect on microphysics in convective cloud is considered (see more discussion in Section 4).

### 2.2. Nudge Technique

Nudge technique is applied to reduce the natural variability of model and to constrain both PD and PI simulations toward the same meteorological conditions (Kooperman et al., 2012). The implementation of nudging in CAM5.3 follows Zhang et al. (2014), in which atmospheric horizontal winds are nudged but temperature is not nudged. The nudged wind fields are taken from the ERA-Interim reanalysis. With this approach, the meteorology is well-constrained while the atmospheric temperature and cloud physics are allowed to rapidly adjust to the forcing (Forster et al., 2016). The simulated mean climate state is not strongly perturbed in model. Briefly, we have
The nudge technique relaxes the model toward a specified time-dependent dynamical state (Telford et al., 2008). The nudging term (Equations 1a and 1b) is applied as part of the “physics” tendency in CAM5.3, where $U$ and $V$ stand for zonal and longitudinal wind components, respectively. Subscript $M$ refers to the model predicted value and subscript $P$ indicates the prescribed variable from the ERA-Interim reanalysis. The relaxation time parameter ($\tau$) determines how tightly the model is constrained with the prescribed conditions and a relaxation time of 6 h is typically used in model studies (Forster et al., 2016; Kooperman et al., 2012; Zhang et al., 2014). Note that we have also examined nudging to the simulated meteorological field, but the main results (e.g., contrasting aerosol effects on LWCF in South East Asia and Amazon) are similar no matter which data are used, and, thus, only the result nudged by ERA-Interim reanalysis is discussed in this study.

2.3. Experimental Design

In this study, we investigate the long-wave ERFari and ERFaci. The LWCF is defined as the difference in net long-wave radiative fluxes at TOA between the cloudy-sky and the clear-sky conditions in CAM5.3. The aerosol effect on LWCF is represented as $\Delta$LWCF. The simulated $\Delta$LWCF includes the instantaneous and subsequent changes in LWCF induced by the increasing aerosols from PI to PD. A series of experiments are designed to isolate the aerosol effects on microphysical, thermodynamic and radiative processes in cloud, and their respective impacts on $\Delta$LWCF (hereafter decomposition experiments). First, we save the aerosol optical properties, the activated CCN and the nucleated IN as outputs in each time step of the PI run, which are subsequently used to estimate the aerosol effect due to Rapid Adjustment to RFari (PI RA), aerosol effect on clouds by acting as CCN (PI CCN), and aerosol effect on cirrus clouds by acting as IN (PI IN) under the PI conditions. Second, we conduct the PD run and rerun it by including each decomposed PI aerosol effect at each time (Table 1). For example, PD(PI RA) is a PD run reading the saved PI values to replace the simulated aerosol optical properties in RRTMG at each model time step. Third, the simulated $\Delta$LWCF from PD-PD(PI RA) run results from the instantaneous change in radiation and subsequent change in cloud properties due to the rapid adjustment to RFari of anthropogenic aerosols increased from PI to PD. Similarly, PD-PD(PI CCN) and PD-PD(PI IN) indicate that the $\Delta$LWCF is attributed to the aerosol effects on clouds by acting as CCN and IN, respectively. In addition to the three individual aerosol effects, a residual effect accounts for the nonlinear interaction of the three aforementioned aerosol effects, and is denoted as PD(PI ALL)-PI, where PD(PI ALL) refers a PD run with the saved PI values of the simulated aerosol optical properties, the activated CCN and nucleated IN at each model time step. In this way, model produces $\Delta$PI $\leftrightarrow$ PD LWCF (see Table 1).

### Table 1

| Experiment name     | Decomposers                                      | Description                                                                 |
|---------------------|--------------------------------------------------|----------------------------------------------------------------------------|
| Total aerosol effect| $\Delta$LWCF                                    | $\Delta$LWCF caused by all aerosol effects from PI to PD                   |
| RA effect           | $\Delta$PI $\leftrightarrow$ PD LWCF (RA effect) | $\Delta$LWCF caused by the rapid adjustment to RFari only                  |
| IN effect           | $\Delta$PI $\leftrightarrow$ PD LWCF (IN effect) | $\Delta$LWCF caused by the aerosol effect on cirrus cloud (aerosols act as IN) only |
| CCN effect          | $\Delta$PI $\leftrightarrow$ PD LWCF (CCN effect) | $\Delta$LWCF caused by the aerosol effect on warm cloud (aerosols act as CCN) only |
| Residual effect     | $\Delta$PI $\leftrightarrow$ PD LWCF (residual effect) | $\Delta$LWCF caused by the nonlinear interaction of the three above aerosol effects |

Abbreviations: CCN, cloud condensation nuclei; IN, ice nuclei; LWCF, long-wave cloud forcing.
However, inserting output from PI run into PD run (one-direction, forward run) could possibly decorrelate some physical connections between aerosol and cloud properties (decorrelation problem) in PD run, making the signal in decomposing runs noisy (see Figure S1 in this manuscript and Appendix of Mulmenstadt et al. [2019]). Therefore, we repeat the above three steps but save output in PD run and insert into PI run, obtaining ∆PI \rightarrow PD LWCF (reversed-direction, backward run, see Table S1). Finally, we apply ∆PI \rightarrow PD LWCF (the arithmetic mean value of ∆PI \rightarrow PD LWCF and ∆PI \leftarrow PD LWCF, two-direction mean result) for analysis in this study, to minimize the noisy signal caused by decorrelation problem. This method has been verified for producing reasonable results for decomposing aerosol effective radiative forcing (Appendix of Mulmenstadt et al. [2019]). Note that we have also conducted the same-climate-different-weather experiments, proving that the forward-backward decomposing method can efficiently alleviate the decorrelation problem in this study (Text S1 and Figure S2). A summarized description of the decomposition experiments is given in Table 1.

### 3. Results and Analysis

#### 3.1. Results

The global ∆LWCF simulated with CAM5.3 is 0.32 W/m² (Figure 1a), which is consistent with the estimate in Kooperman et al. (2012) and within the range of estimates in Ghan et al. (2012) (note that ∆LWCF defined in this study corresponds to the sum of LW indirect and semi-direct effects in Ghan et al. [2012]). The global ∆LWCF is not as significant as the aerosol effect on SWCF in CAM5 (about 1/5 to 1/3 in magnitude as in Kooperman et al. [2012], Zhang et al. ([2014], and in this study). The simulated ∆LWCF is specifically prominent in two tropical regions (Figure 1a): South East Asia (the region within 10°S–30°N and 60°E–120°E, abbreviated as SEA hereafter) and Amazon (the region within 20°S-10°N and 40°W-80°W, as AMZ hereafter). This is physically reasonable because cirrus clouds occur in higher levels (with lower cloud top pressure, see Figure S3) in tropics and higher clouds have stronger effects on long-wave warming. Aerosols in both SEA and AMZ regions are plentiful due to biomass burning and anthropogenic sources. However, ∆LWCF exhibits contrasting patterns (Figure 1a) in SEA (warming effect) and in AMZ (cooling effect), whereas the shortwave effect shows concurrent cooling in these two regions (Figure 2a). Expanded high cloud fraction and elevated cloud top from PI to PD are responsible for the reduction of outgoing long-wave radiation and the resulted warming in SEA, while the cooling in AMZ is caused by decreased cloud fraction in high level and lower cloud top (Figures 2a and 3a). The model estimated ∆LWCF in CAM 5.3 is +2.43 W/m² in SEA and −1.96 W/m² in AMZ (Tables 2 and 3).

### Table 2

|          | ∆LWCF (W/m²) | SEA | AMZ |
|----------|--------------|-----|-----|
| ANM      | 2.43         | −1.96 |
| DJF      | 1.73         | −2.08 |
| MAM      | 1.31         | −2.54 |
| JJA      | 4.80         | −1.01 |
| SON      | 1.89         | −2.19 |

Abbreviation: LWCF, long-wave cloud forcing.

### Table 3

|          | ∆LWCF (W/m²) | ∆CLDHGH (%) | ∆CTP (hPa) | ∆LWP (g/m²) | ∆IWP (g/m²) | ∆R-ice (micron) |
|----------|--------------|-------------|------------|-------------|-------------|-----------------|
| SEA      | AMZ          | SEA         | AMZ        | SEA         | AMZ         | SEA             | AMZ             |
| Total aerosol effect | 2.43   | −1.96       | 1.64       | −2.67       | −12.14      | 18.2            | 16.23           | 10.99           | 0.21          | −1.68         | −1.25         | −0.13         |
| RA effect | −0.12     | −0.16       | 0.05       | −0.24       | −1.22       | 2.82            | −0.68           | −0.33           | −0.11         | −0.13         | 0.04          | 0.012         |
| IN effect | 1.92        | −0.47       | 1.75       | −0.74       | −12.05      | 4.79            | 0.76            | −0.22           | 0.55          | −0.21         | −1.14         | 0.23          |
| CCN effect | 0.43       | −1.46       | −0.29      | −2.02       | 1.29        | 12.99           | 14.66           | 10.3            | −0.29        | −1.29         | −0.21         | −0.13         |
| Residual effect | 0.26     | −0.23       | 0.31       | −0.25       | −2.47       | 1.46            | 1.22            | 0.75            | 0.098        | −0.18         | 0.018         | 0.016         |

Note. CLDHGH, high cloud fraction; CTP, cloud top pressure; LWP, cloud liquid water path; IWP, ice water path; R-ice, ice particle radius; ∆ is the changes from PI to PD in the run with total aerosol effect, but represents ∆PI \rightarrow PD in the four decomposing runs for RA, IN, CCN, and residual effects. Abbreviations: CNN, cloud condensation nuclei; IN, ice nuclei; PD, present day; PI, preindustrial.
Results from the decomposition experiments clearly indicate that the positive $\Delta$LWCF in SEA is attributed mainly to the IN effect whereas the negative $\Delta$LWCF in AMZ is due to the CCN effect (Figures 1c and 1d and Table 3). The RA effect has negligible impacts on the changes of LWCF, high cloud fraction and cloud top height (Figures 1b, 2b, and 3b). The increased cloud top ($-12.05$ hPa) and extended high cloud fraction ($+1.75\%$) with increased aerosols are dominant in SEA (Figures 2c and 3c and Table 3) when IN effect is considered alone. On the contrast, the decreased cloud top ($+12.99$ hPa) and shrunk high cloud fraction ($-2.02\%$) with increased aerosols are dominant in AMZ due to CCN effect (Figures 2d and 3d and Table 3).

Note that the simulated ice cloud amount and the cloud top pressure show good agreements with the retrievals from Clouds and the Earth’s Radiant Energy System-Moderate Resolution Imaging Spectroradiometer (Figure S3), lending confidence to the validity of modeled cloud properties. In addition, different ice
nucleation schemes (BN and KL schemes) in CAM5.3 exhibit similar pattern of contrasting ∆LWCF in SEA and in AMZ as the result in Figure 1a with LP scheme, but with smaller magnitudes (Figure 4).

3.2. Further Analysis

3.2.1. CCN Effect

This subsection focuses on the mechanisms of ∆LWCF resulting from aerosol effect on warm clouds by acting as CCN. Figure 5 shows the changes in CCN and IN concentration from PI to PD in tropics. On the one hand, the increases of activated CCNs mostly occur in low-level warm clouds and peaks in SEA (90 E–120 E) and AMZ (60 W–90 W) in tropics (Figure 5a). The enhanced sulfate and carbonaceous aerosols are responsible for the increasing CCN globally, especially in East Asia, South Asia, Europe, Eastern North America, and Amazon. On the other hand, the increases of nucleated INs mostly occur above 200 hPa and peak in SEA (60 E–120 E) in tropics (Figure 5b). The change in the nucleated IN from PI to PD has a similar
pattern with the change in water vapor amount in ice phase cloud layer (Figures 6a and 6b). But the sulfate IN for homogeneous nucleation and dust IN for heterogeneous nucleation have no obvious changes in AMZ and in SEA ocean areas, only SEA land areas have a reduced sulfate IN from PI to PD (Figure 6). These results suggest that the change of cloud top height/pressure in AMZ and in SEA is more likely influenced by some other factors (e.g., water vapor, see the analysis afterwards) in addition to aerosols acting as CCN or IN in CAM5.3.

The ice cloud fraction in tropics is mainly occupied by stratiform clouds in high levels with negligible contribution of the simulated tropical convective clouds (the convective cloud occupies the cloud fraction mostly in low and mid-levels below 500 hPa [Figures S5a and S6a]). Therefore, the aerosol effect on cirrus cloud is mainly relevant to the interactions between aerosol and large-scale stratiform clouds.
The ice stratus fraction in CAM5.3 is diagnosed by a quadratic function of grid mean RH over ice. The value of ice RH is regulated by moisture content and ice cloud water content from the previous model time step (Park et al., 2014). As a result, both vertical transport of water vapor and ice cloud microphysics are critical to the ice cloud fraction. Figure 7b plots the variation of vertical velocity in tropics from PI to PD. The tropical upward motion is clearly suppressed around 100°E (within SEA region) and 75°W (within AMZ region), where the activated CCNs are increased due to anthropogenic aerosols (Figure 5a). The global pattern of increased CCN is also consistent with the enhancement of cloud water path from PI to PD (Figures 8b and 8e). These are evidences for the cloud albedo effect and lifetime effect (ERFacI) and could be major reasons for the suppressed vertical motion in SEA and AMZ (Figures 7 and 8). As we know, more numerous CCN in warm cloud results in smaller cloud droplet size (not shown) and inhibits the collision-coalescence process, thus, reducing the precipitation efficiency and enhancing the cloud water path. Clouds with higher water path are optically thicker, leading to more reflected shortwave radiation and less transmitted radiation reaching the surface. Therefore, the surface is cooled (Figures S4b and S4e) and vertical motion is suppressed. The weakened upward motion and reduced vertical transport of water vapor disfavor the ice cloud formation.

The CCN effect can also be seen from the clear water vapor divergence in the land regions of SEA and AMZ at 859 hPa (Figure 9d), which results from the weakened upward motion. The local humidity decreases with the divergence of water vapor in SEA. In the ocean area of SEA, compensatory convergence of water vapor occurs and water vapor can be transported to the high levels by upward motion. That is why the
strengthened upward motions at 80 E and 130 E accompany the weakened motion at 100 E in SEA (Figure 7e). Similar water vapor convergence can be found in oceans around AMZ (Figure 9d).

Among the different aerosol effects, CCN effect is mostly responsible for the weakening of vertical motion (Figures 7b and 7e and Table 3). RA effect, IN effect, and residual effect have trivial influences on cloud water path (Figures 8c, 8d, and 8f) and upward motion (Figures 7c, 7d, and 7f). In land regions of SEA and in AMZ, the enhanced cloud water path in low levels cools the surface and suppresses the vertical motion, resulting in a negative ΔLWCF (long-wave cooling effect) together with the reduced cloud fraction in high level and lower cloud top.

3.2.2. IN Effect

Aerosol effect on ice phase microphysics could potentially influence the development of ice cloud and thus the LWCF. Figure 10b shows the change in effective radius of ice crystals from PI to PD. Reduction of 1–2 micron in ice particle size mainly occurs around 60 E–120 E (mostly ocean areas in SEA) in the tropical region, the location with the increasing nucleated IN (Figure 5b). Among the different aerosol effects, IN effect is specifically responsible for the change of ice particle size (Figure 10d and Table 3) in high levels (layers with temperature below −37°C), which indicates that the aerosol effect on ice phase microphysics is relevant to the homogeneous freezing instead of the heterogeneous nucleation at the layers below −37°C in CAM5.3 (Figure S7 in this manuscript and Figure 4a in Gettelman et al. [2012]). The smaller ice particles have smaller falling speed and maintain the occurrence of ice clouds by expanding the ice cloud coverage or developing to the higher altitude (Figures 2c and 3c). The enhanced ice cloud fraction and cloud top height result in a positive ΔLWCF (long-wave warming effect).
Figure 7. Distribution of vertical velocity (Pa/s) in tropical regions (averaged over 30°S-30°N) in (a) PD run. Changes of tropical vertical velocity in (b) PD-PI run (with all aerosol effects), in decomposing runs for (c) RA effect, (d) IN effect, (e) CCN effect, and (f) residual effect. CCN, cloud condensation nuclei; IN, ice nuclei; PD, present day; PI, preindustrial.

Figure 11 further illustrates the aerosol effects on ice particle number and mass concentrations over land and ocean regions of the SEA. In the land of SEA and AMZ, CCN effect (yellow curves) tends to reduce the ice particle number and ice water content (Figures 11a, 11c, 11d, and 11f) due to the weakened upward motion caused by ERFaci on warm clouds as analyzed in Section 3.2.1. In the ocean region of SEA, both number and mass of cloud ice particles are promoted by CCN effect (yellow curves in Figures 11b and 11e).
due to the water vapor convergence (Section 3.2.1). Furthermore, IN effect (green curves) tends to increase the number but reduce the size of ice crystals, thus, prolonging the existence of ice clouds in both land and ocean of SEA. RA effect (blue curves) has negligible impact on the changes of cloud ice comparing to the other aerosol effects.

3.2.3. Summary

Figure 12 illustrates the different mechanisms responsible for the contrasting \( \Delta \text{LWCF} \) in SEA and in AMZ as simulated in CAM5.3. Comparing to the clean condition in PI simulation (leftmost cloud in Figure 12), the increased aerosols in PD simulation act as CCNs and ACI inhibits the development of ice cloud, resulting in the reduced ice cloud fraction and cloud top height (rightmost cloud comparing to the leftmost one in Figure 12) and a long-wave cooling effect (negative \( \Delta \text{LWCF} \)). The mechanism of CCN effect dominates in AMZ and in the land of SEA. However, IN effect overwhelms in the ocean of SEA. From PI to PD, the
increased aerosols act as homogeneous IN and reduce the ice particle size, prolonging the existence of ice clouds in high altitude (cloud in the middle comparing to the leftmost one in Figure 12) and resulting in a long-wave warming effect (positive ∆LWCF).

In CAM5.3, the aerosol effects on LWCF are related to not only the microphysics, but also the thermodynamic and radiative processes. Therefore, the seasonal variation of clouds will affect the strength of ∆LWCF. Table 2 lists the simulated ∆LWCF in the four seasons of SEA and AMZ. In northern hemispheric summer, cloud fraction is high in SEA (Figure S8), thus, the long-wave warming (+4.80 W/m²) is strong, while the cloud fraction in AMZ is relatively small, causing a weaker long-wave cooling (−1.01 W/m²). In addition, summer monsoon transports abundant water vapor to SEA, making ice clouds more sensitive to the increases of aerosols. On the opposite, summer in the northern hemisphere is the dry season in AMZ, thus, constraining the aerosol effect on clouds with the limited water vapor. Finally, comparative analysis (Table 3) shows that in SEA, IN effect is the largest contributor to the positive ∆LWCF (+1.92 W/m²), followed with CCN effect (+0.43 W/m²), whereas CCN effect dominates all the other effects for the negative ∆LWCF (−1.46 W/m²) in AMZ.

4. Concluding Remarks

A series of numerical experiments are conducted with CAM5.3 to dissect the various aerosol effects on LWCF; contrasting results are found in the tropical regions of South East Asia and Amazon. Our analysis indicates that the cooling (negative ∆LWCF) in Amazon is due mainly to the aerosol effect on warm clouds and the inhibition of vertical motion by the aerosol-induced radiative cooling. In contrast, the warming (positive ∆LWCF) in South East Asia is due mainly to the aerosol effect on ice nucleations, especially for the homogeneous freezing, thus, reducing the ice particle size and prolonging the existence of ice cloud. Our results emphasize that a comprehensive analysis of integrated aerosol effects on both warm and ice clouds is necessary for better understanding the change of LWCF.
Several points are noteworthy. First, this study is based on the CAM5.3 only. Simulations with only one climate model and the experimental design have several constraints. Different climate models could generate different global patterns of \( \Delta \text{LWCF} \) due to the different dynamic frames, thermodynamic or physical parameterizations. For example, ECHAM6 (the sixth generation of ECHAM) with KL scheme and GEOS5 (the Goddard Earth Observing System, Version 5) with BN scheme produced negative \( \Delta \text{LWCF} \) over Amazon and positive \( \Delta \text{LWCF} \) over Indian Ocean and Pacific Ocean around South East Asia, similar as the current study. But these two models generate strong \( \Delta \text{LWCF} \) in mid-latitude oceanic and continental areas with...
comparable strength to that in tropical areas (unpublished results via personal communication with Dr. Xiaohong Liu, simulated ∆LWCF from ECHAM6 and GEOS5, were collected under the framework of the Aerosol Comparisons between Observations and Models [AEROCOM] phase III experiments), while the ∆LWCF in mid-latitudes is much weaker than that in AMZ and SEA in this study (Figure 1a). In addition, simulations with the Multiscale Aerosol Climate Model (MACM) in Kooperman et al. (2012) produced a negative global mean ∆LWCF, in contrast to the overall positive ∆LWCF in Figure 1a. MACM has extended

![Figure 11. Vertical profiles of the changes in number concentration and mass mixing ratio of ice particles (NI and QI) in the respective decomposition experiments over land and ocean of South East Asia (SEA, the left two columns) and in Amazon (AMZ, the right column). Black: PD-PI run (with all aerosol effects), blue: decomposing run for RA effect, green: decomposing run for IN effect, yellow: decomposing run for CCN effect, and red: decomposing run for residual effect](image)

![Figure 12. A schematic diagram to illustrate the different mechanisms of contrasting long-wave cloud forcing due to aerosol effects](image)
the multiscale modeling framework approach to include the enhanced aerosol physics contained in CAM5. Convection was resolved so that aerosols and convective clouds were connected in MACM, but aerosol particle concentrations are not directly linked to ice nucleation. MACM generated weaker aerosol effects on LWCF than CAM5, which resulted from both smaller changes in aerosol burden and a weaker relationship between CCN and liquid water content (Kooperman et al., 2012). Therefore, the aerosol effects on LWCF could be highly dependent on the selection of atmospheric model, aerosol module, and ACI schemes.

Second, the combination of different ice nucleation schemes could strongly affect the competition of water vapor between homogeneous freezing and heterogeneous ice nucleation. Incorporation of soot as heterogeneous IN may add further uncertainty in the cloud forcing caused by increasing anthropogenic aerosols (Gettelman et al., 2012).

Third, it is expected that the aerosol effect on the development of convective cloud could have impacts on high level cloud fraction, cloud top height and a resulted impact on LWCF. Future study is merited to couple ACI parameterizations with the convective parameterization in current GCM (e.g., Song et al., 2012).

Finally, the nudged meteorological field could potentially buffer the aerosol-induced effects. On the one hand, the aerosol impact on the meteorology is more or less reduced by nudging the meteorological field every 6 h. On the other hand, the emission amounts of natural aerosols in PI and PD runs were constrained by nudging to the same meteorological field (winds).

### Author Contributions

M. Wang ran the model simulation and analyzed the results. Y. Peng designed the research and wrote the manuscript. Y. Liu helped with the experimental design and analysis.

### Data Availability Statement

The source code for CAM5.3 is distributed through a public Subversion code repository. This code can be checked out using Subversion client software, such as the command tool `svn`, or simply view the version with a web browser https://svn-ccsm-models.cgd.ucar.edu/cesm1/release_tags/cesm1_2_1/. The input data necessary to run CAM5.3 are made available at https://svn-ccsm-inputdata.cgd.ucar.edu/trunk/inputdata/. Model results plotted and analyzed in this study are available at https://doi.org/10.5281/zenodo.4262357.

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