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To cite this article: Yi-Chun Chen *et al* 2020 *Environ. Res. Lett.* **15** 034047

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Environmental Research Letters



LETTER

OPEN ACCESS

RECEIVED

11 August 2019

REVISED

30 January 2020

ACCEPTED FOR PUBLICATION

11 February 2020

PUBLISHED

5 March 2020

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Evaluation of sea salt aerosols in climate systems: global climate modeling and observation-based analyses*

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Abstract

Sea salt aerosols (SSA), one of the most abundant aerosol species over the global oceans, play important roles for Earth's climate. State-of-the-art SSA parameterizations in global climate models (GCMs) are typically modeled using near-surface wind speed, sea surface temperature (SST), and precipitation. However, these have non-trivial biases in CMIP3 and CMIP5 GCMs over the tropical Pacific Ocean that can contribute to biases in the simulated SSA. This study investigates the impacts of falling ice radiative effects on the biases of the aforementioned modeled parameters and the resulting modeled SSA biases. We compare the CMIP5 modeled SSA against satellite observations from MISR and MODIS using a pair of sensitivity experiments with falling ice radiative effects on and off in the CESM1-CAM5 model. The results show that when falling ice radiative effects are not taken into account, models have weaker surface wind speeds, warmer SSTs, excessive precipitation, and diluted sea surface salinity (SSS) over the Pacific trade-wind regions, leading to underestimated SSA. In the tropical Pacific Ocean, the inclusion of falling ice radiative effects leads to improvements in the modeled near-surface wind speeds, SSTs, and precipitation through cloud-precipitation-radiation-circulation coupling, which results in more representative patterns of SSA and reduces the SSA biases by ~10% to 15% relative to the satellite observations. Models including falling ice radiative effects in CMIP5 produce smaller biases in SSA than those without falling ice radiative effects. We suggest that one of the causes of these biases is likely the failure to account for falling ice radiative effects, and these biases in turn affect the direct and indirect effects of SSA in the GCMs.

1. Introduction

Despite decades of research, aerosols and clouds continue to contribute to large uncertainties in estimates of the Earth's changing energy budget (Boucher *et al* 2013). To reduce these uncertainties, it is essential for atmospheric models to realistically represent the geographical distributions of aerosols; this, in turn, is critical for accurate simulation of Earth's surface energy and water budgets. However, the present-day representation of

aerosols and clouds and, more specifically, their interactions in general circulation models (GCMs), are major contributors to the remaining uncertainties in climate change predictions (Seinfeld *et al* 2016).

Sea spray aerosol including sea salt aerosols (SSA), the largest natural aerosol source, is produced over more than 70% of the Earth's surface and plays an important role in the Earth's radiative budget (Seinfeld and Pandis 2006). SSA perturb the radiation budget through scattering and absorption of solar radiation and emitting thermal infrared radiation (referred to as aerosol direct effects). Observations in Nakajima *et al* (2001) showed there is a positive correlation between

* The data that support the findings of this study are available from the corresponding author upon reasonable request.

Table 1. List of coupled atmosphere-ocean climate models in CMIP5 archive historical run used in this study and corresponding institute.

| Model | Institute |
|-----------|---|
| CESM1 | National Center for Atmospheric Research (NCAR, USA) |
| HadGEM2 | Met Office Hadley Center ESM (UK) |
| BNU-ESM | College of Global Change and Earth System Science, Beijing Normal University (China) |
| IPSL-CM5 | Institute Pierre-Simon Laplace (France) |
| MIROC-ESM | Japan Agency for Marine-Earth Science and Technology, Atmosphere and Ocean Research Institute (The University of Tokyo), and National Institute for Environmental Studies (Japan) |
| MRI-CGCM3 | Meteorological Research Institute (Japan) |

cloud optical thickness and oceanic aerosol number concentration, whereas the effective particle radius has a negative correlation with aerosol number concentration. Many studies have suggested SSA are the main sources of cloud condensation nuclei over the ocean (e.g. Pierce and Adams 2006), affecting the microphysical properties of shallow planetary boundary layer marine clouds and hence their radiative properties, amount, and lifetime (referred to as aerosol indirect effects). Aerosols are coupled with clouds through aerosol indirect effects and semi-direct effects (IPCC AR5). Changes in aerosol abundance affect the response of trade-wind shallow cumulus, including cloud microphysical and macrophysical properties (e.g. Xue *et al* 2008, Saleeby *et al* 2015), convection (e.g. Jiang *et al* 2018), precipitation (e.g. Kogan *et al* 2012), and cloud lifetime (e.g. Jiang *et al* 2006). Significant discrepancies in aerosol optical depth (AOD) between GCMs and satellite observations contribute to biases in estimating aerosol direct and indirect radiative forcing, and therefore the climate feedback.

Over the ocean, the main emission of SSA is through air bubbles bursting at the air-sea interface, meaning the production of SSA is directly affected by air-sea interactions such as near surface winds, sea surface wind stress, sea surface temperature (SST), sea surface salinity (SSS), and precipitation (Woodcock 1953, Seinfeld and Pandis 2006). Most parameterizations in GCMs assume that SSA emission has a strong dependence on 10 m wind speed (Monahan *et al* 1986). Some parameterizations also include dependencies on different meteorological parameters, such as SST (Mårtensson *et al* 2003, Jaeglé *et al* 2011, Witek *et al* 2016) and SSS (Ovadnevaite *et al* 2014).

In the Community Atmosphere Model version 5 (CAM5) of the Community Earth System Model version 1 (CESM1), the parameterization of SSA sources is derived from a method proposed by Mårtensson *et al* (2003), which is based on lab experiment for particles with dry diameters (D_p) from 20 to ~ 2500 nm. The particle flux, F , as a function of whitecap area is expressed as:

$$\frac{dF}{d \log D_p} = W \times (A \times SST + B), \quad (1)$$

where $W = 3.84 \times 10^{-4} U_{10}^{3.41}$, U_{10} is the wind speed at 10 m above the sea surface; A and B are size dependent polynomials, with coefficients listed in table 1 of

Mårtensson *et al* (2003). For dry diameters $> 2-3 \mu\text{m}$, the parameterization is based on Monahan *et al* (1986), derived as $\frac{dF}{d \log D_p} = C \times U_{10}^{3.41}$, where C is a function of particle size.

Other than SSA produced by breaking waves, the depletion of SSA includes wet deposition and dry deposition (gravity and turbulence). As the parameterizations are dependent on 10 m wind speed, SST, and precipitation, etc, model biases caused by these parameters via other dynamical and physical errors can lead to errors in the modeled SSA indirectly. One cause of the aforementioned model biases could be attributed to missing representation of the radiative effects of falling ice (snow or precipitating ice) radiative effects in the models (Li *et al* 2015; supplementary information, available online at stacks.iop.org/ERL/15/034047/mmedia). Li *et al* (2015) pointed out that all the GCMs in the Coupled Model Intercomparison Project phase 3 (CMIP3) and most in the phase 5 (CMIP5) do not consider precipitating ice radiative effects, and these models tend to have too strong convection, producing anomalous low-level outflow (see SI, figure S2(b)) over the Intertropical Convergence Zone (ITCZ), South Pacific Convergence Zone (SPCZ), and Maritime Continent (See SI, figure S3). This leads to weakening surface wind stress, weaker low-level winds and upper ocean mixing, resulting in warmer SSTs (See SI, figure S4), producing excessive precipitation, and diluting SSS over trade-wind regions (Li *et al* 2014, 2016, 2018). The following question will be addressed in this study: to what extent do the responses (i.e. weaker surface winds, warmer SSTs, and excessive precipitation) from lacking falling ice radiative effects contribute to the GCM biases in SSA simulations? In other words, what are the impacts on local SSA through the remote influence of changes in circulation? The SSA biases would in turn affect the shallow convection and aerosol direct effects, and thus climate feedback.

In this study, we explore the impacts of missing falling ice radiation interactions on aerosol loadings parameterized in equation (1) for SSA by conducting a pair of sensitivity tests by turning on or off falling ice radiative effects in CESM1-CAM5, and comparing the results to aerosol data from the Multi-angle Imaging SpectroRadiometer (MISR) and MODerate-resolution Imaging Spectroradiometer (MODIS) satellite

instruments. This paper is structured as follows: section 2 includes a brief description of the model simulations and key observations used. Section 3 describes the biases in CMIP5 reports compared to observations and results from sensitivity experiments. Conclusions and a discussion of major findings are provided in section 4.

2. Model and data description

2.1. Model and sensitivity experiment

The National Center for Atmospheric Research—Department of Energy (NCAR-DOE) CESM1 is a coupled climate model for simulating the Earth's climate system (for details see section 2 in Li *et al* (2015)). The descriptions and performance of the cloud-related physical parameterizations in its atmosphere component, CAM5, can be found in Morrison and Gettelman (2008), Gettelman *et al* (2010), and Lindvall *et al* (2013).

In this study, numerical experiments were performed with the diagnostic falling ice radiative effects turned off (hereafter, NoS) and on (hereafter, S). The changes of model fields due to the inclusion of falling ice radiative effects are calculated as the difference between the simulation without and with these effects, which is referred to as 'NoS-S.' Both simulations are set up in the same manner as used in CMIP5. The specific scenario used in these sensitivity experiments is the CMIP5 historical runs (1850–2005) with exactly the same model components used in the CESM1 contribution to CMIP5, where the same initial fields from preindustrial and the observed 20th century greenhouse gas, ozone, aerosol, and solar forcing are used (Taylor *et al* 2012). The simulation time period used in the analyses presented here is 1970–2005.

2.2. Satellite AOD data

In this study, AOD observations from MISR and MODIS are utilized. MISR was launched in December 1999 onboard Terra, NASA's flagship Earth Observing System (EOS) spacecraft. MISR consists of nine cameras that view the Earth simultaneously at different angles (nadir and 26.1°, 45.6°, 60.0°, and 70.5° forward and backward of nadir) and observes each scene over an interval of 7 min with a common swath of approximately 400 km (Diner *et al* 1998). Global coverage is achieved every nine days. MISR's standard, operational data products are available and listed in the following link: http://eosweb.larc.nasa.gov/project/misr/misr_table.

MODIS aboard NASA's Terra satellite, has 36 spectral bands with wavelengths from 0.41 to 14 μm and its 2330 km wide swath provides global coverage every two days. Both MODIS and MISR have been widely used for monitoring aerosol properties and both provide long time series of global aerosol abundances and properties. In this study, we utilize Level 3 monthly gridded data from MISR Version 22 and

MODIS Collection 6.1 Dark Target (DT) to compare with model results.

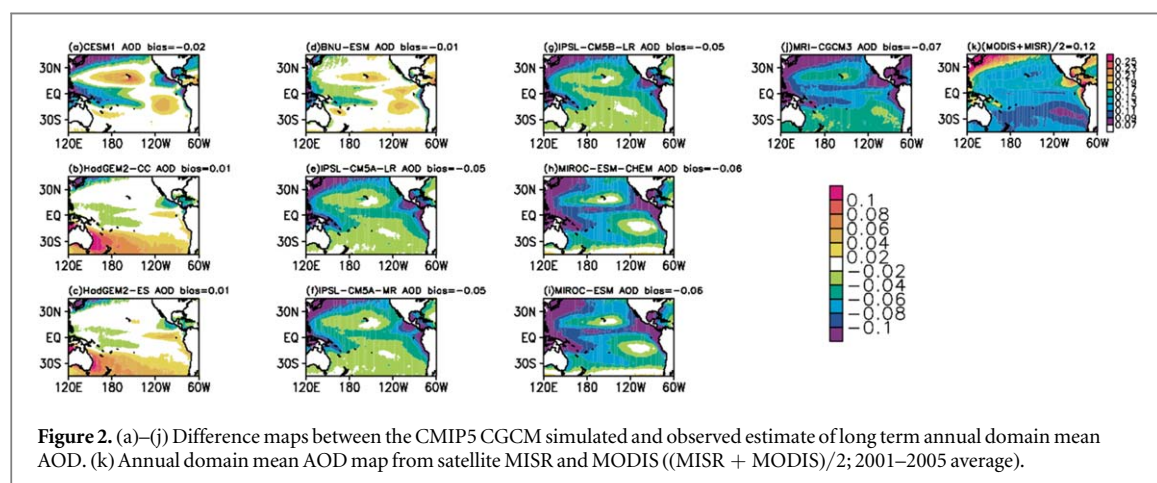
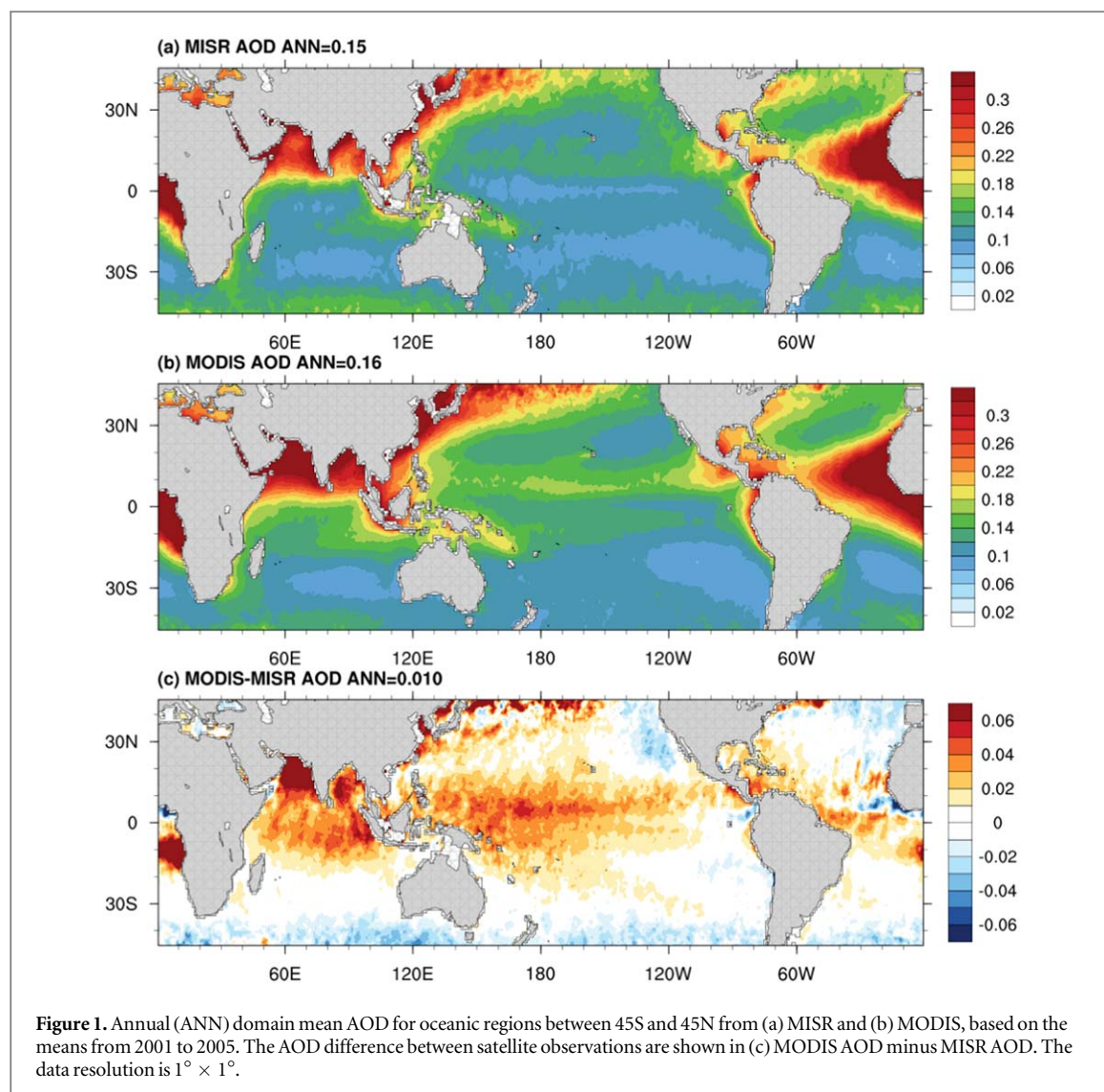
Figure 1 shows the annual mean AOD distribution over the global ocean from these satellite observations based on the time period from 2001 through 2005. The spatial pattern and magnitudes are similar between the MISR and MODIS annual AODs (figures 1(a) and (b)), with a mean AOD difference of about 0.01 over global ocean (figure 1(c)). Based on Kahn *et al* (2009), the MISR-MODIS correlation coefficient is about 0.9 over ocean.

For ocean, the MODIS Collection 6.1 DT AODs agree well with the Aerosol Robotic Network (AERONET) AODs (correlation coefficient = 0.880, and root-mean-square error = 0.083) with 73.5% of the data samples falling within the expected errors (as shown in figure 3(a) in Wei *et al* 2019). The uncertainty of MODIS AOD is $\pm(0.03 + 5\% \times \text{AOD})$ over ocean (Remer *et al* 2005, Wei *et al* 2019). As for MISR, about 70%–75% of MISR-retrieved AOD values falls within the greater of 0.05 or $20\% \times \text{AOD}$ of AERONET (Kahn *et al* 2010). The correlation coefficients between MISR AOD and AERONET are ~ 0.9 for maritime stations (Kahn *et al* 2010). In the version of the product used here, the retrieval of MISR AOD over water may be subject to several known issues, including cloud contamination of retrieved AOD, camera artifacts in high-contrast scenes, residual calibration effects, etc (e.g. Witek *et al* 2018a, 2018b). Despite different sources of uncertainty from the two satellite retrievals, these data sets are the best available in terms of their long term records and global coverage, and figure 1(c) provides confidence that their performance is similar over global oceans. For model and observations comparisons, we use the long-term annual mean AOD output from 10 CMIP5 CGCM historical runs (1970–2005) with the mean annual AOD retrieved from MISR and MODIS (2001–2005) (see figure 2).

3. Model results

3.1. CMIP5 model AOD bias

Figures 2(a)–(j) show the biases of long-term mean (1970–2005) spatial distribution of AOD in the Pacific from 10 CMIP5 CGCMs (see table 1) as compared with satellite observations, shown in figure 2(k) for reference. For six of the models, the simulated annual mean AOD is underestimated relative to mean satellite AOD, while four of the models (figures 2(a)–(d)) show relatively good agreement. For the three models in which falling ice radiative effects are considered (figures 2(a)–(c)), the AOD bias is relatively small over the tropical Pacific with a mean of -0.002 (figure 3(a)). In comparison, the AOD over the tropical Pacific is significantly underestimated by those models that do not consider falling ice radiative effects (figures 2(e)–(j)), which overall have a mean bias of -0.047 (figure 3(b)).



It has already been demonstrated that non-trivial biases exist in those CMIP3 and CMIP5 models that do not have falling ice radiative effects, with the resulting simulations exhibiting excessive precipitation, too-weak low-level winds, and underestimated SSS over Pacific trade-wind regions (Li *et al* 2014, 2016, 2018). Here we investigate how these biases affect the

simulated AOD through sensitivity studies performed using CESM1 as described in the following section.

3.2. Sensitivity results from falling ice radiative effects using CESM1

Using a pair of sensitivity tests turning on or off falling ice radiative effects in CESM1-CAM5, we examine the

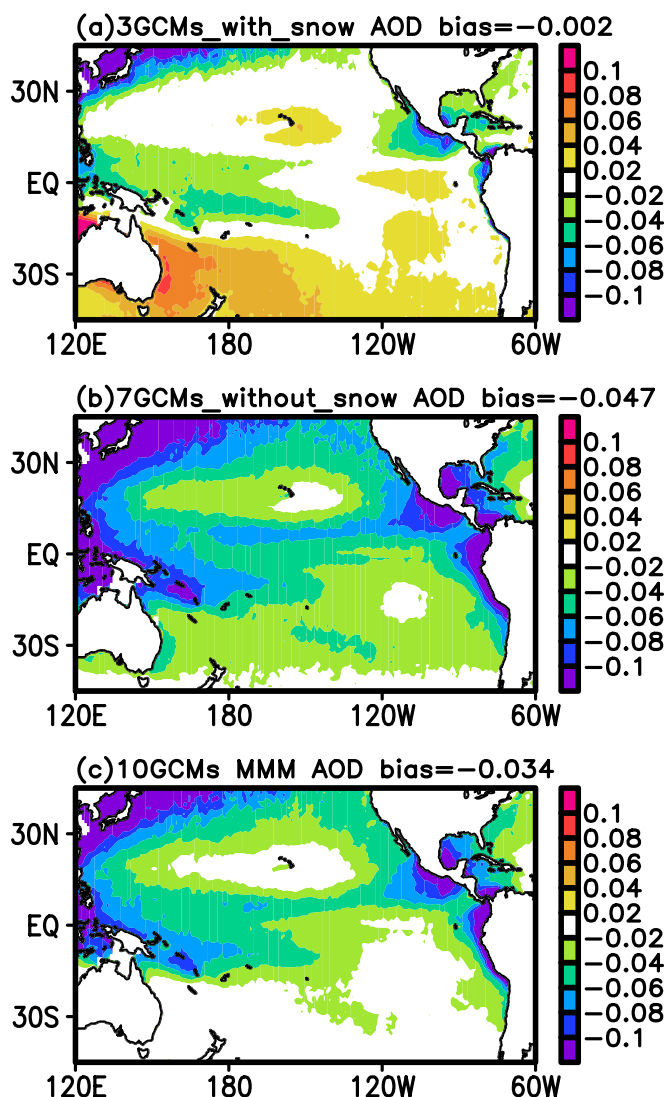
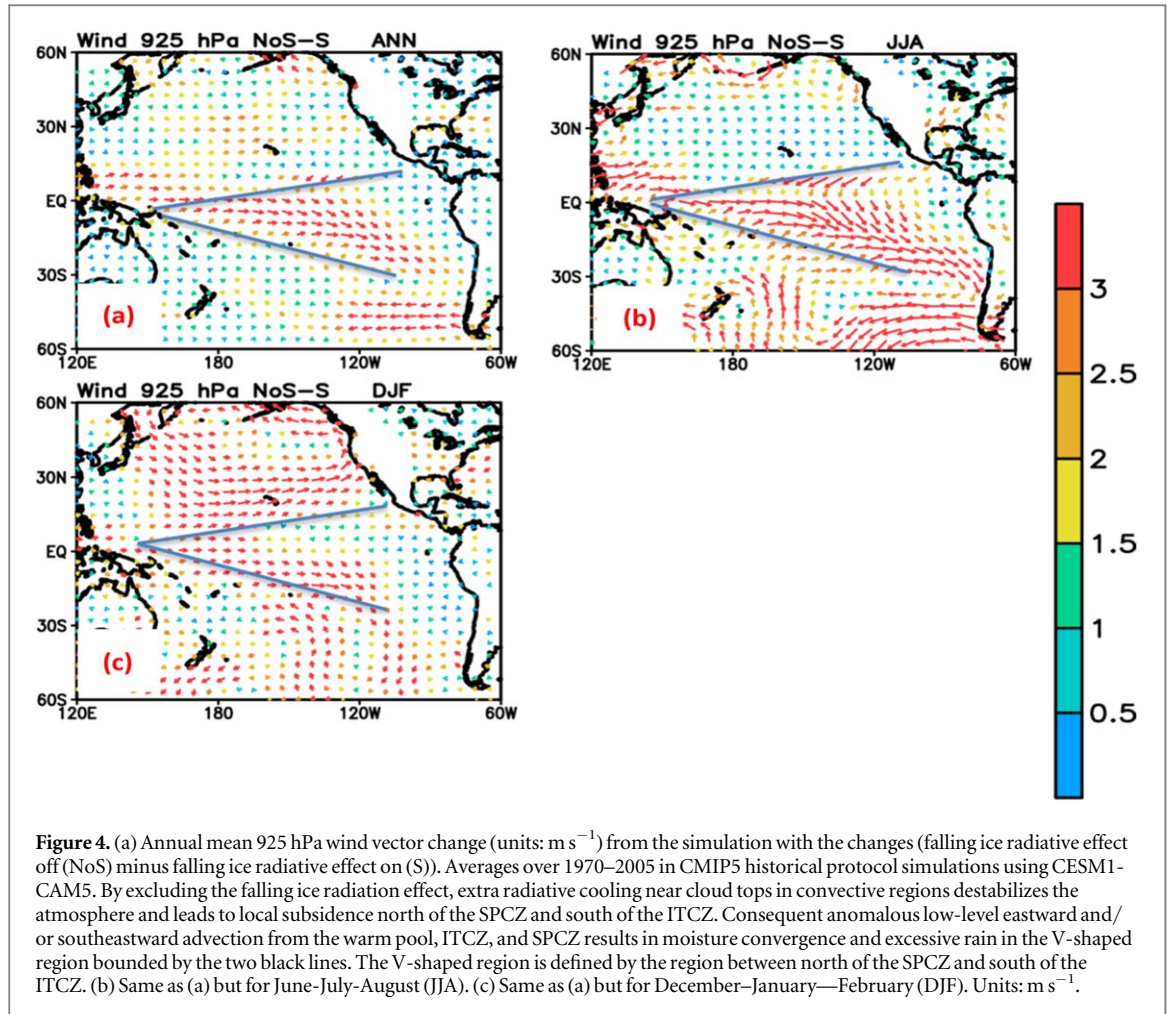


Figure 3. (a) Mean AOD bias from 3 GCMs with falling ice radiative effects (i.e. CESM1_S, HadGEM2-CC, and HadGEM2-ES) as compared to satellite observations from MODIS and MISR. (b) same as (a), but from 7 GCMs without falling ice radiative effects (i.e. models from figures 2(d)–(j)). (c) same as (a), but from all 10 GCMs.

impacts of falling ice radiative effects on the SSA bias. Based on the parameterization of SSA generation in CESM1 represented in equation (1), the sensitivities of wind speed, SST, and precipitation to the falling ice radiative effects are examined over Pacific trade-wind regions, in particular.

Figure 4 shows the 925 hPa wind differences for 1970–2005 for the NoS-S scenarios from our CESM1-CAM5 simulations. The V-shaped marking in figure 4 highlights a region where an anomalous southeast low-level outflow is clearly found over the northern warm pool and southern Pacific trade wind regions in the annual mean state (figure 4(a)). The smallest wind speed differences between the NoS and S scenarios are observed in the boreal summer months (JJA, figure 4(b)) while the largest wind speed differences are observed in boreal winter (DJF, figure 4(c)). These patterns are also common in the CMIP5 models that lack falling ice-radiation effects (see SI).

The differences in annual means of AOD, 10 m wind speed, precipitation, and SST between NoS and S (NoS-S) for the Pacific basin are shown in figure 5. The AOD difference between NoS and S simulations (figure 5(a)) is more apparent from the Western Pacific warm pool to the north of the SPCZ, with lower AOD in NoS simulations (those not including falling ice-radiation interactions). In the NoS case, relative to the S case, there are weaker easterly trade winds (figure 5(b)) caused by low-level outflow, resulting in weaker upper ocean mixing along with excessive precipitation (figure 5(c)), and warmer SST (T_s ; shown in figure 5(d)) over the tropical Pacific with underestimated AOD. This demonstrates there are systematic biases in the coupling of the precipitation-radiation-circulation caused by the lack of falling ice radiative effects simulated in CESM1. Through changes in wind, precipitation, and SST, the abundance of SSA is altered via dynamical air-sea coupling processes.



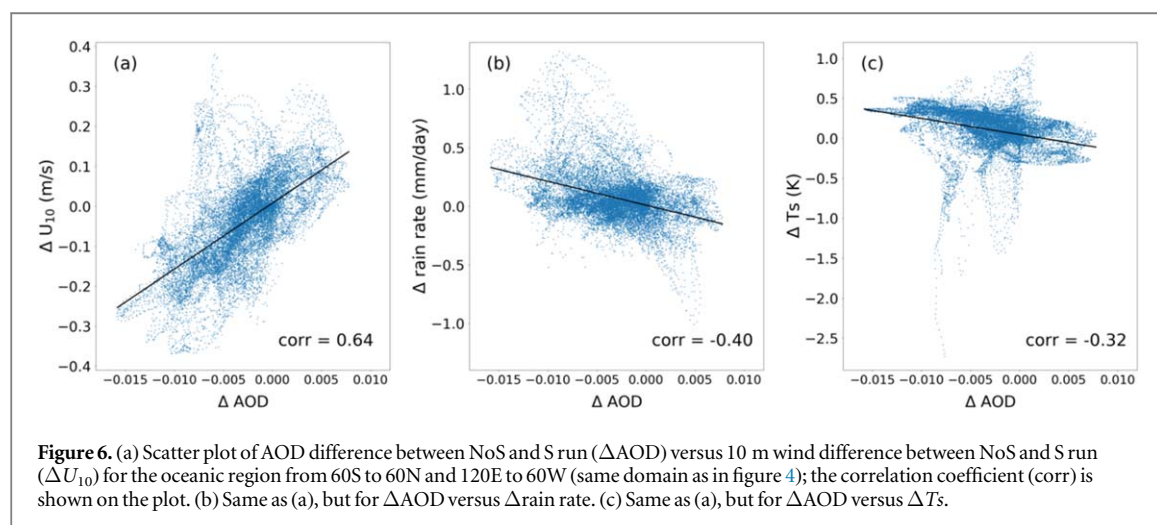
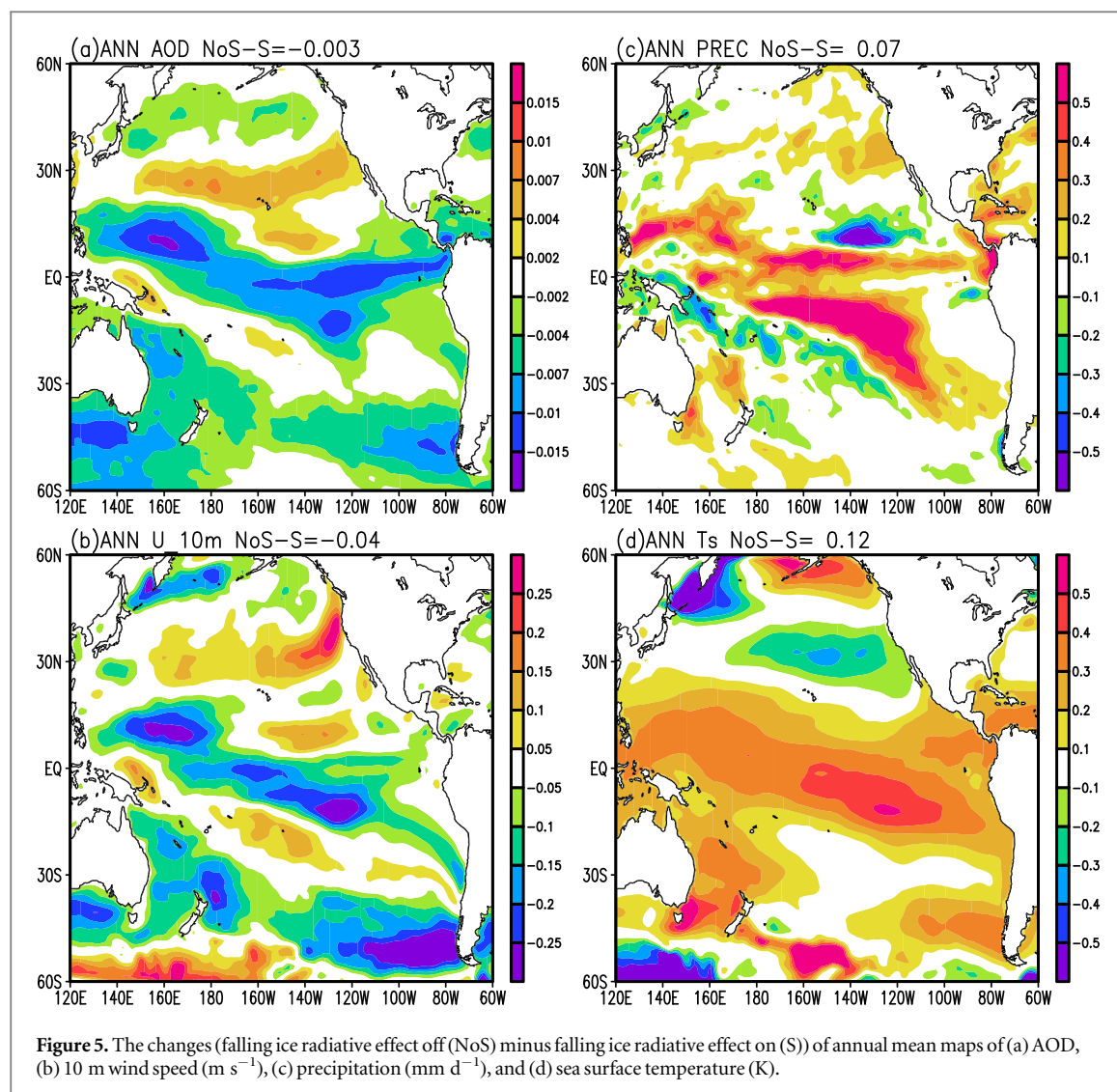
The correlation in the Pacific between the difference in AOD and the change in each dependent parameter was examined. The AOD difference between NoS and S is highly correlated with the difference in wind speed (figure 6(a)), with a domain-averaged spatial correlation coefficient of 0.64. This is not surprising, as the source of SSA depends on wind speed to the power of 3.41, as shown in equation (1). On the other hand, excessive precipitation leads to a negative correlation between the change in precipitation and the change in AOD from the NoS and S simulations, with a domain-averaged correlation coefficient of -0.40 (figure 6(b)). For AOD and SST, the correlation coefficient is -0.32 (figure 6(c)), which indicates a somewhat weaker relationship compared to AOD-wind speed and AOD-precipitation. The relationship between AOD and SST is dependent on the size of the aerosol particles according to laboratory experiments carried out by Mårtensson *et al* (2003). For aerosols with dry diameters $<0.07 \mu\text{m}$ ($>0.35 \mu\text{m}$), the number concentration decreases (increases) with increasing (decreasing) water temperature. For the size range from 0.07 to $0.35 \mu\text{m}$, there is no clear temperature dependence of the aerosol concentrations. The correlations between AOD difference and precipitation/SST difference (-0.4 and -0.32) are not robust enough, which might be due to the offset contributions from the 10m wind speed (positive

contribution) and precipitation/SSTs (negative contribution) to the net AOD changes.

Figure 7 shows the improvement from NoS to S against the annual mean AOD from MISR and MODIS observations. The satellite data are from 2001 to 2005, corresponding to the period of overlap between model simulations and observations. As shown in figure 2(a), comparing the CESM1 simulations with MISR and MODIS, the simulated AODs are underestimated in the western central Pacific Ocean near the equator and in the northwest Pacific Ocean. In order to determine whether the inclusion of falling ice radiation interactions can decrease the biases between the model and the observations, the absolute improvement from NoS to S run compared to observations is calculated as:

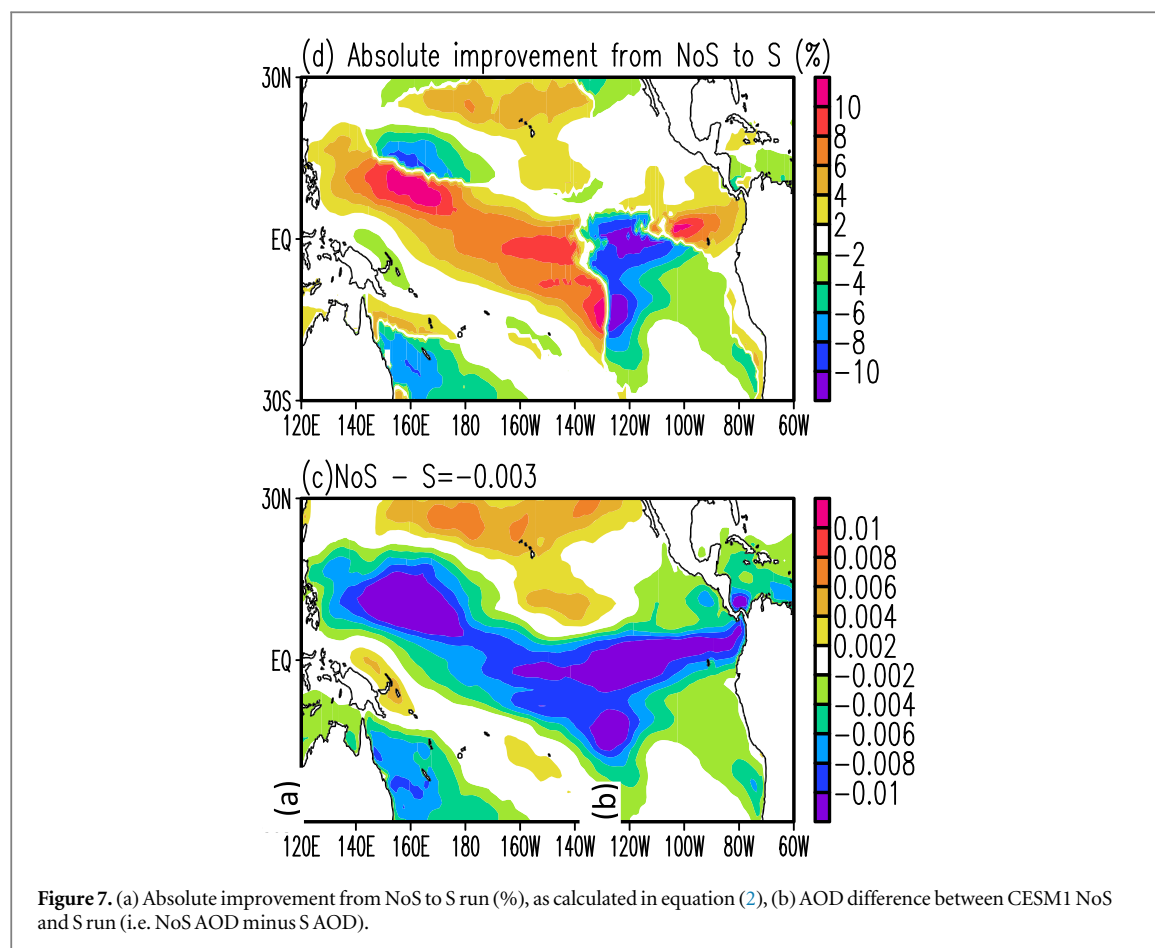
$$\begin{aligned} \text{Absolute improvement (\%)} &= \left(\left| \frac{AOD_{\text{NoS}} - AOD_{\text{satellite}}}{AOD_{\text{satellite}}} \right| - \left| \frac{AOD_{\text{S}} - AOD_{\text{satellite}}}{AOD_{\text{satellite}}} \right| \right) \times 100\%. \quad (2) \end{aligned}$$

Positive values indicate improvement from NoS to S. In figure 7(a), regions with positive values (reddish



colors) indicate improvement when the falling ice radiative effects are included in the CESM1 run, whereas regions with negative values (darker colors) indicate worse agreement. Improvements are found in most areas of the Pacific Ocean, except for certain regions in the eastern central Pacific and southwest

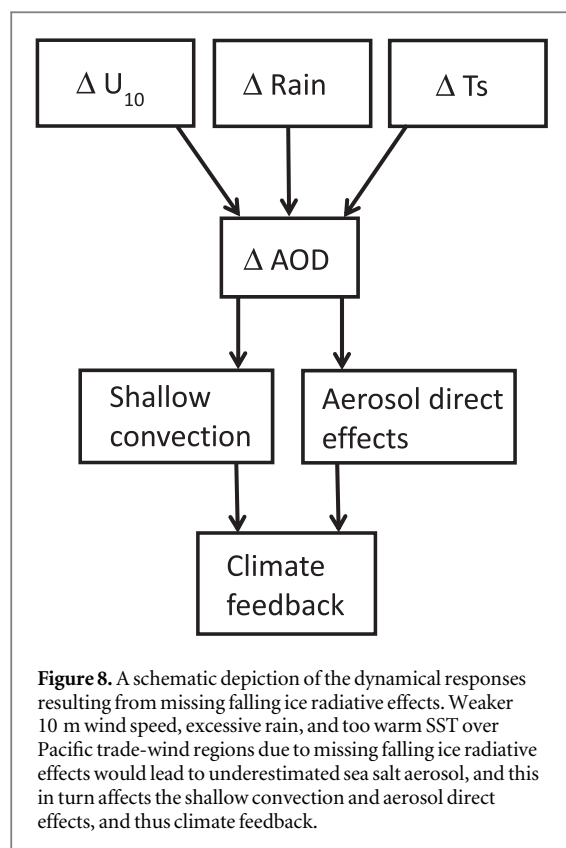
Pacific near Australia. The largest improvement occurs in the central Pacific. With the falling ice radiative effects included, simulated AOD in the central Pacific Ocean improves by $\sim 10\%$ to 15% . The patterns are more or less matched with the difference between NoS and S shown in figure 7(b).



4. Conclusion and discussion

Oceanic aerosols are important for supplying cloud condensation nuclei, and affect Earth's climate energy and water budget through their coupling to clouds via the indirect and semi-direct effects. In GCMs, the representation of SSA is dependent upon modeled near-surface wind speed, SST, and precipitation. However, most GCMs do not consider falling ice radiative effects, resulting in too-warm SSTs, weaker surface winds and excessive precipitation over the Pacific trade-wind regions. To explore whether these effects might lead to underestimated SSA, we examined the impacts of falling ice radiative effects on modeled SSA using CESM1-CAM5 by turning on and off falling ice radiative effects (S and NoS). We found that when neglecting the falling ice radiative effects in CESM1, the model produces weaker low-level winds, excessive precipitation, and warmer SST, leading to underestimated SSA through precipitation-cloud-circulation coupling. The above mentioned biases are commonly found in most CMIP5 models. The geographic distribution and the values of the SSA bias are found to be mainly controlled by the modeled biases in precipitation and surface wind speeds. With the inclusion of falling ice radiation effects in CESM1, the SSA absolute biases decrease by $\sim 10\%$ to 15% in the central Pacific Ocean.

We conclude that the exclusion of falling ice radiative effects in most GCMs partially contributes to the modeled AOD biases. We found that in most CMIP5 models the differences between the NoS and S scenarios correspond to anomalously weaker outflow in the western Pacific warm pool and southern Pacific trade wind regions, which reduces upper ocean mixing and ultimately increases SST and precipitation and decreases sea surface salinity. It is suggested that one cause of bias between modeled SSA and satellite data is the missing representation of the falling ice radiative effects in the models. As indicated schematically in figure 8, the local SSA can be affected by local and non-local air-sea coupling. The SSA bias could be reduced by improving the model-simulated near surface winds, SST, and precipitation. For GCMs including the falling ice radiative effects, the SSA biases have decreased, as shown in CESM1-CAM5 and two HadGEM2 models in CMIP5. We found that the bias in the simulating SSA radiative effects can therefore be influenced by the biases either locally or remotely determined by other parameters, in this case, precipitation, surface winds, and SSTs through air-sea coupling. We suggest that when dealing with aerosol biases in model, other than tuning the emissions, consideration of the dynamical processes affecting aerosol abundance is also essential.



Acknowledgments

YCC acknowledge RCEC, Academia Sinica for supporting this study. The authors acknowledge Marcin Witek for his review and comments. The contribution by JLL, DJD, MJG, JHJ, and OVK to this study was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under contracts of ATMOS COMP 2013 (NNH12ZDA001N-CCST) with the National Aeronautics and Space Administration (NASA). This work has been supported in part by the NASA Making Earth System Data Records for Use in Research Environments (MEaSUREs) programs. The authors acknowledge PCMDI/DOE and the WCRP's WGCM for their roles in making CMIP3/CMIP5 available. MODIS aerosol data were downloaded using the GES-DISC Interactive Online Visualization and Analysis Infrastructure, a part of the NASA's Goddard Earth Sciences Data and Information Services Center. MISR aerosol data (MIL3MAE.004) were obtained from the NASA Langley Research Center Atmospheric Science Data center (ASDC).

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