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Global distribution and climate forcing of marine organic aerosol – Part 2: Effects on cloud properties and radiative forcing

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Abstract

In the first part of this paper series (Meskhidze et al., 2011), a treatment of marine organic aerosols (including primary organic aerosol, secondary organic aerosols, and methane sulfonate) was implemented into the Community Atmosphere Model version 5 (CAM5) with a 7-mode Modal Aerosol Module. A series of simulations was conducted to quantify the changes in aerosol and cloud condensation nuclei concentrations in the marine boundary layer. In this study, changes in the cloud microphysical properties and radiative forcing resulting from marine organic aerosols are assessed. Model simulations show that the anthropogenic aerosol indirect forcing (AIF) predicted by CAM5 is decreased in absolute magnitude by up to ∼0.10 W m^{-2} (8 %) when marine organic aerosols are included. Changes in the AIF from marine organic aerosols are associated with small global increases in low-level in-cloud droplet number concentration and liquid water path of ∼1.3 cm^{-3} (∼1.6 %) and 0.2 g m^{-2} (0.5 %), respectively. Areas especially sensitive to changes in cloud properties due to marine organic aerosol include the Southern Ocean, North Pacific Ocean, and North Atlantic Ocean, all of which are characterized by high marine organic emission rates. As climate models are particularly sensitive to the background aerosol concentration, this small but non-negligible change in the AIF due to marine organic aerosols provides a notable link for ocean-ecosystem marine low-level cloud interactions and may be a candidate for consideration in future earth system models.

1 Introduction

Marine organic aerosols, emitted into the atmosphere as primary particles via bursting of bubbles at the ocean surface and secondary particles via oxidation of volatile organic compounds (VOC) such as isoprene, monoterpenes, amines, and dimethyl sulfide (DMS), have been shown to affect the chemistry and number distribution of aerosols in the marine environment (O'Dowd et al., 2004; Meskhidze and Nenes, 2006;

Yoon et al., 2007; Facchini et al., 2008). Part 1 of this study (Meskhidze et al., 2011) described the implementation of marine organic aerosols in the Community Atmosphere Model version 5 (CAM5) with a 7-mode Modal Aerosol Module (MAM-7) (Liu et al., 2011). Meskhidze et al. (2011) showed that addition of marine organics led to improved agreement of the model predicted and measured concentrations of organic aerosols in the marine boundary layer and increased the cloud condensation nuclei (CCN) concentrations by up to 20 % over biologically active oceanic regions. Here in the second part of the study, we focus on the impact of the marine organic aerosols on cloud microphysical properties and shortwave radiative forcing.

2 Model experimental setup

To detect the specific effects of marine organics on aerosol and cloud properties, various sensitivity simulations expanding upon the simulations in Meskhidze et al. (2011) have been performed. In the Part 1 simulations, the "Default" simulation in which there are no marine organic aerosols was compared to simulations which included production of marine secondary organic aerosol (SOA) and methane sulfonate (MS^-) and/or marine POA emissions from two schemes (Vignati et al., 2010; Gantt et al., 2011) that were either externally- or internally-mixed. A consistent set of emissions are used in the sensitivity simulations for this work, including the Gantt et al. (2011) marine primary organic aerosol (POA) emissions (selected due to their resulting concentrations having a more favorable comparison to measurements than that of Vignati et al., 2010) and production of marine SOA and MS^- which together are hereinafter referred as "G11". Model simulations are carried out with the G11 emissions to examine sensitivity to different aerosol activation parameterizations, marine POA hygroscopicity and mixing state treatment, and the magnitude of anthropogenic emissions. Tables 1a, b give a summary of the 5-yr long simulations with 3-month spin-up, including those performed in Part 1 on which simulations in this work are based. Detailed descriptions of the different aerosol activation schemes can be found in Meskhidze et al. (2011) and thus will not be repeated here. An additional sensitivity test was conducted for the hygroscopicity parameter (κ) (Petters and Kreidenweis, 2007) of marine POA by increasing it (in the G11 simulation) from the default $\kappa = 10^{-10}$ value used for terrestrial POA to $\kappa = 0.1$ (Liu et al., 2011). This adjusted κ represents the upper end of the potential marine POA hygroscopicity based on the measurements of κ values of 0.006 and 0.04 for estuarine (Moore et al., 2008) and riverine (Svenningsson et al., 2006) organic matter, respectively. In order to estimate the effect of marine organic aerosols on cloud radiative forcing, both the Default and G11 simulations were performed with present-day (PD) and pre-industrial (PI) aerosol and precursor emissions. The anthropogenic aerosol indirect forcing (AIF) is then calculated as the difference in model-predicted short wave cloud forcing (SWCF) between PD and PI conditions. The model simulations with PD and PI emissions used anthropogenic emissions from the IPCC AR5 dataset for the year 2000 and 1850, respectively (Bond et al., 2007; Junker and Liousse, 2008; Lamarque et al., 2010).

3 Results

As shown in Meskhidze et al. (2011), treating marine organic aerosols increased the simulated aerosol mass and CCN concentration over much of the ocean. Here we expand upon these results by carrying out a number of sensitivity tests for additional model parameters with the emphasis on the changes in cloud microphysics and radiative forcing associated with marine organic aerosols. Unless specified otherwise, the reported changes are relative to the Default simulation.

3.1 Effects on cloud physical properties

3.1.1 Low-level cloud droplet number

With the addition of CCN from marine organic aerosols shown in Meskhidze et al. (2011), a resulting increase in the in-cloud droplet number concentration (CDNC)

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is expected assuming a constant liquid water path. This effect should be particularly pronounced for low clouds as marine aerosols are typically found within 1 km above the ocean surface (Kiliyanpilakkil and Meskhidze, 2011). Table 2 shows that compared to the Default simulation, the global annual mean increase in low-level CDNC (between 945 and 980 mb) from marine organics in the G11 simulation is 1.3 cm^{-3} (∼1.6 %) while over the ocean CDNC increases by ∼1.4 cm^{-3} (∼2.2 %). The spatial distribution of low-level CDNC from the Default and G11 simulations shown in Fig. 1a and 1b reveals that the increase occurs mostly over the Southern Ocean and Northern Atlantic. Many of these areas also have the greatest percentage changes (up to 20 %) in CDNC as shown in Fig. 1c and are statistically significant (with a p-value <0.1 according to the paired t-test in Fig. S1). Figure 1c also shows some areas with decreases in low-level CDNC; these changes are typically not statistically significant (see Fig. S1), and are likely due to model noise. Changes (absolute difference as opposed to percentage difference) in low-level CDNC due to marine OA show a seasonal dependence similar to that of [Chl-*a*] at some higher latitude areas. The spatial distribution of low-level CDNC percentage changes is similar to the percentage changes in the surface concentration of CCN at 0.2 % supersaturation shown in Meskhidze et al. (2011). The magnitude of low-level CDNC changes in our simulations is much lower than the increases of up to ∼300 % predicted in Roelofs (2008) for the Northern Atlantic Ocean; this difference is likely due to the much higher submicron marine organic aerosol number concentration in Roelofs (2008) resulting from higher emission rates (Roelofs (2008) has up to 25 Tg C yr^{-1} of emissions in both the Aitken and accumulation modes while G11 emits 0.1 and 2.1 Tg C yr^{-1} in the marine POA Aitken and accumulation modes, respectively).

3.1.2 Column cloud properties and radiative forcing

In addition to increases in low-level CDNC, marine organic aerosols in CAM5 lead to changes in the grid-cell averaged column CDNC, liquid water path (LWP), and SWCF. Like the low-level CDNC, the column CDNC (see Fig. 2a) experiences statistically significant (Fig. S1) increases of up to 20 % over the Southern Ocean where emissions are the highest. Globally, the percentage increases in column droplet number from the Default to the G11 simulation of 0.4 % (1.1 % over the ocean) is smaller than that of low-level CDNC (see Table 2) as marine organic aerosol typically remain within the boundary layer. For the LWP, Fig 2b shows that despite the seemingly random regions of positive and negative changes between the Default and G11 simulations, there are widespread areas over the Southern Ocean that experience a large (up to 20 %) increase in the LWP as a result of marine organic aerosols. The spatial distribution of the changes in SWCF shown in Fig. 2c is similar to that of LWP with roughly 10 % decreases (~ $-5\,\mathrm{W\,m^{-2}}$) in the vicinity of Falkland (52° S, 58° W) and South Georgia Islands (54° S, 36° W) in the South Atlantic Ocean where emissions of marine organic aerosols are very high. Note that the SWCF has a negative value; therefore decrease in SWCF indicates less solar radiation reaching the surface and shows as a positive percentage change in Fig. 2c. The spatial location of this large decrease in SWCF is similar to Meskhidze and Nenes (2006) who reported a satellite-derived $15\,\mathrm{W\,m^{-2}}$ decrease in TOA short-wave radiation due to changes in the properties of liquid clouds over a summertime phytoplankton bloom near South Georgia Island. The discrepancies in magnitude are likely to be associated with the coarser model grid, annual averaging (summertime SWCF changes of -5 to $-10\,\mathrm{W\,m^{-2}}$ are predicted throughout the region), and selection by Meskhidze and Nenes (2006) of a time period with a particularly large phytoplankton bloom. Globally, the changes in the LWP and SWCF are much smaller relative to the region near South Georgia Island. Table 2 shows that there is a $0.2\,\mathrm{g\,m^{-2}}$ (0.5 %) global average increase in the LWP in the G11 simulation relative to the Default simulation, and a $0.1\,\mathrm{W\,m^{-2}}$ (0.2 %) decrease (indicating the increased reflection of shortwave radiation) in the SWCF. Like low-level CDNC, there is a seasonality in the absolute differences of column CDNC, LWP, and SWCF between the G11 and Default simulations at some high latitude areas that is similar to the [Chl-*a*] seasonality.

3.2 Sensitivity simulations

3.2.1 Aerosol activation parameterizations

Table 2 shows that the two activation schemes, Abdul-Razzak and Ghan (2000) scheme (referred to as AR-G) that is used in all but two of the simulations, and the Fountoukis and Nenes (2005) (referred to as FN) give considerably different global low-level CDNC, LWP, and SWCF values (Default vs. Default-FN). Ghan et al. (2011) showed that these differences produce a $0.2\,\mathrm{W\,m^{-2}}$ (10 %) smaller anthropogenic AIF with the FN scheme ($-1.60\,\mathrm{W\,m^{-2}}$ with FN and $-1.76\,\mathrm{W\,m^{-2}}$ with AR-G). However, the changes due to marine organic aerosols are relatively consistent between the two schemes (Default vs. G11 and Default-FN vs. G11-FN), suggesting that the variability in aerosol activation parameterizations does not alter the net climatic impact of marine organic aerosols. The global increases of $1.2\,\mathrm{cm^{-3}}$, $0.2\,\mathrm{g\,m^{-2}}$, and decreases of $0.1\,\mathrm{W\,m^{-2}}$, for the low-level CDNC, LWP, and SWCF, respectively, between the Default-FN and the G11-FN simulations are comparable to the changes ($1.3\,\mathrm{cm^{-3}}$, $0.2\,\mathrm{g\,m^{-2}}$, and $-0.1\,\mathrm{W\,m^{-2}}$, for the low-level CDNC, LWP, and SWCF) between the Default and G11 simulations that use the AR-G scheme.

3.2.2 Marine POA mixing state

In Meskhidze et al. (2011), the changes in surface CCN concentration at 0.2 % supersaturation were shown to be quite sensitive to the mixing state of marine POA emissions, with an external mixture (added marine POA mass emissions are accompanied by corresponding increases to sea spray number emissions) yielding a much greater effect on CCN number compared to an internal mixture (only the sea spray aerosol mass is enhanced by addition of organics). Table 2 shows that relative to the G11 simulation, which includes an external mixture of marine POA emissions, the simulation with an internal mixture of marine POA emissions (G11-Internal) has consistently lower CDNC and LWP and higher (lower absolute magnitude) SWCF. Meskhidze et al. (2011) showed that when marine organics were added as an internal mixture with sea-salt the model predicted a slight reduction in CCN concentration, even over biologically productive waters of the Southern Ocean. In such a case the increase in CCN number due to the growth of mean modal diameter (caused by the addition of organic mass) is outweighed by the decrease in particle hygroscopicity. As both external and internal mixtures of marine organic aerosols and sea salt have been observed (Hultin et al., 2010), these mixing state differences likely represent upper and lower estimates for the effect of marine organic aerosol on cloud microphysical properties and resulting shortwave radiative forcing.

3.2.3 Hygroscopicity

A change in value of κ for marine POA from 10^{-10} used for G11 to 0.1 (as was used for terrestrial POA in the sensitivity tests of Liu and Wang, 2010) is expected to yield more CCN at relevant supersaturations and hence a greater impact on cloud properties and radiative forcing. However, Table 2 shows that the global and annual mean values from the G11 simulation and the G11-κ (the simulation identical to G11 but with marine POA κ increased to 0.1) reveal this not to be the case; model predicted low-level CDNC, LWP, and SWCF in the G11-κ simulation are nearly equivalent to those of the G11 simulation. There are two likely reasons for this insensitivity to aerosol hygroscopicity: (1) the aging method of aerosols in CAM5 based on a criterion of 3 mono-layers of sulfate, and (2) the hygroscopicity-dependent wet removal rates in CAM5 (Liu et al., 2011). Non-seasalt sulfate concentrations within the marine boundary layer are relatively high (>0.2 µg m^{-3}) over productive waters where most of the marine POA emissions are coincident with strong DMS emissions; this causes the modeled aerosol aging in these areas to be more rapid than in oceanic areas without strong DMS emissions. As a result, cloud properties and SWCF are not sensitive to the hygroscopicity of the freshly-emitted marine POA, but rather to the κ value of aged marine POA which is the same in G11 and G11-κ. This insensitivity to the POA hygroscopicity due to sufficient aging by sulfate has been observed previously in internally-mixed particles (as

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are simulated by CAM5), particularly over the industrial regions (Chang et al., 2007; Prenni et al., 2007; Wang et al., 2008; Liu and Wang, 2010; Liu et al., 2011). Furthermore, the atmospheric lifetime and burden of marine POA have an inverse relationship with hygroscopicity because more hygroscopic aerosols experience wet scavenging before those with low hygroscopicity (Liu et al., 2011). As a result, the increase in CCN (at a given supersaturation) due to higher hygroscopicity is offset somewhat by the decrease due to more rapid wet removal.

3.2.4 Preindustrial and present day anthropogenic emissions

It has been well documented that increased anthropogenic emissions since the beginning of the industrial revolution have an impact on the present day aerosol burden and associated cloud microphysical properties. Here we examine how the climate forcing due to changes between PI and PD emissions may be influenced by marine organic aerosols. In this set of simulations, all parameters are held constant except for the anthropogenic aerosol and precursor emissions: the Default and G11 simulations use emissions for the year 2000, while the Default-PI and G11-PI simulations use emissions for the year 1850. Recent studies suggest considerable decline (more than 6 %) in ocean primary productivity over the past decade likely attributed to anthropogenic activities and associated climate change (Bopp et al., 2001; Gregg et al., 2003; Behrenfeld et al., 2006). However, the difficulties in separating the natural variability from the global climate change trend has also been proposed (Henson et al., 2010). Due to the absence of reliable ocean productivity data for PI conditions, we use the same monthly chlorophyll-*a* concentrations (effectively resulting in the same primary and precursor emissions for marine organic aerosols) for both G11 and G11-PI simulations. Table 2 shows that the addition of marine organics has a larger impact on cloud properties for simulations with PI emissions (differences between G11-PI and Default-PI) compared to the simulations with PD emissions (differences between G11 and Default). The global and annual mean differences in PI low-level CDNC, LWP, and SWCF are 1.3 cm^{-3} (2.3 %), 0.3 $g\,m^{-2}$ (0.7 %) and -0.2 $W\,m^{-2}$ (0.5 %), respectively,

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while the mean differences over the ocean are 1.7 cm^{-3} (3.6 %), 0.4 $g m^{-2}$ (1.2 %) and −0.2 $W m^{-2}$ (0.5 %), respectively. The difference in the climatic effect of marine organic aerosol with and without high levels of anthropogenic aerosols is due to the higher susceptibility of preindustrial clouds to additional aerosols (Platnick and Twomey, 1994; Lohmann and Lesins, 2002). Specifically, these simulations show that additional CCN from marine organic aerosol emissions affect the properties of clouds in clean regions more than those in polluted regions. This result is consistent with the study by Hoose et al. (2009) where the slope of the change in SWCF between PI and PD emissions decreases with increases in the prescribed aerosol concentration.

4 Aerosol indirect forcing

The effect of marine organic aerosols on anthropogenic AIF is explored for various model configurations. Calculations show that marine organic aerosols reduce the absolute magnitude of the global average AIF from a default value of −1.35 $W m^{-2}$ to -1.25 $W m^{-2}$ (an 8 % change) when comparing the Default/Default-PI and G11/G11-PI differences. Although Wang et al. (2011) have suggested that the sensitivity of SWCF to changes in aerosol loading may be too strong in CAM5, this is not expected to have considerable influence on the relative effect of marine organic aerosols. This change in AIF by the marine OA (+0.10 $W m^{-2}$) is similar in magnitude to the SWCF changes between the Default and G11 simulations for both PD (−0.11 $W m^{-2}$) and PI (−0.21 $W m^{-2}$). In contrast, the SWCF change between the Default and G11-Internal for PD is a factor of 10 smaller, which suggests a much smaller change to AIF by marine OA if the marine POA emissions are considered to be internally mixed with sea salt. As described in Sect. 3.2.4, the reduction of AIF due to marine organic aerosols mainly occurs due to an increase in background low-level CDNC. This result is consistent with Hoose et al. (2009) which showed lower predictions of changes in SWCF with an increase in the minimum CDNC or aerosol concentration. Figure 3a and b show that oceanic areas like the Southern Ocean and northern Atlantic Ocean, which

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have high marine organic emissions and low/moderate levels of CDNC, experience the largest decrease in the absolute value of AIF when marine organic aerosols are included. In the more heavily polluted oceanic region downwind of China where the AIF is the greatest, marine organic aerosols have little impact. As concentrations of marine organic aerosols are typically well below those of terrestrial and anthropogenic aerosols in all but "clean" marine air masses, it is not surprising that these areas are the most sensitive to their inclusion. Overall, our calculations suggest that the bi-directional feedbacks between marine organic aerosols, clouds and climate can have a non-trivial impact on the cloud albedo effect predicted by climate models (IPCC, 2007). Therefore, the inclusion of marine organic aerosols has the potential to improve model estimates of CCN, cloud microphysics and model-predicted shortwave radiative forcing especially over remote marine regions.

5 Conclusions

A treatment of marine organic aerosols has been implemented into the CAM5 coupled with MAM-7 to examine their effects on cloud properties and radiative forcing. Results show that marine organic aerosols can have large local effects on clouds (up to a 20 % annual average increase in low-level in-cloud CDNC in the Southern Ocean, Northern Pacific and Northern Atlantic), especially during the summertime when chlorophyll-*a* concentration ([Chl-*a*]) is typically at a maximum. When these aerosols are included in model comparisons of pre-industrial (PI) and present day (PD) anthropogenic emissions, the model-predicted absolute value of anthropogenic aerosol indirect forcing (AIF) can be decreased by up to 0.10 $W m^{-2}$ (8 %). Predicted changes in low-level CDNC and shortwave cloud forcing due to marine organic aerosols are more sensitive to the mixing state of the primary marine organic aerosols (internal vs. external mixtures) than their hygroscopicity (chemistry). As both external and internal mixtures of marine organic aerosols and sea-salt have been observed, this sensitivity to mixing state highlights the need for improved understanding of the emission pro-

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cesses and their implementation in global models. The global changes in cloud microphysical and radiative properties due to marine organic aerosols are higher in the simulations with preindustrial [1.3 cm^{-3} (2.3 %) and −0.2 $W m^{-2}$ (0.5 %) for low level CDNC and SWCF, respectively] than present day emissions [1.3 cm^{-3} (1.6 %) and -0.1 $W m^{-2}$ (0.2 %) for low level CDNC and SWCF, respectively]. This result is consistent with studies showing that pristine clouds have the highest susceptibility to increased aerosol concentrations (Platnick and Twomey, 1994). As climate models are sensitive to the background aerosol concentration and CDNC in remote marine areas (Menon et al., 2002), this study demonstrates the importance of accurate prediction of marine aerosol-cloud-climate interactions for future assessments of model-predicted extent of human-induced climate change.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/12/7453/2012/acpd-12-7453-2012-supplement.pdf.

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Table 1a. Description of the CAM5 simulations[a] in the Part 1 paper.

Simulation	Emissions
Default	sulfur dioxide, sulfate, terrestrial POA, terrestrial SOA, black carbon, ammonia, dust, DMS, sea-salt
SOA/MS^-	Same as Default but with marine SOA, MS^-
G11	Same as Default but with Gantt et al. (2011) marine POA emissions externally-mixed with sea-salt, marine SOA, MS^-
V10	Same as Default but with Vignati et al. (2010) marine POA emissions externally-mixed with sea-salt, marine SOA, MS^-
G11-Internal	Same as G11, except marine POA emissions internally-mixed with sea-salt, no marine SOA or MS^-
V10-Internal	Same as V10, except marine POA emissions internally-mixed with sea-salt, no marine SOA or MS^-

[a] All simulations were 5-years long with a 3-month spinup.

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Table 1b. Description of the CAM5 simulations[a] in the Part 2 paper.

Simulation	Emissions	Setup
Default-PI	Same as Default but with anthropogenic emissions set to year 1850	Same as Default
Default-FN	Same as Default	Uses the Fountoukis and Nenes (2005) (FN) aerosol activation parameterization
G11-FN	Same as G11	Uses the (FN) aerosol activation parameterization
G11-PI	Same as G11 but with anthropogenic emissions set to year 1850	Same as G11
G11-κ	Same as G11	Same as G11 but with hygroscopicity value (κ) for marine POA set to 0.1

[a] All simulations were 5-years long with a 3-month spinup

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Table 2. CAM5 modeled mean liquid cloud properties.

Simulation	Low-level CDNC (cm^{-3})[a]		Column CDNC ($\times 10^9\ m^{-2}$)[b]		LWP ($g\,m^{-2}$)[b]		SWCF ($W\,m^{-2}$)	
	Global	Ocean	Global	Ocean	Global	Ocean	Global	Ocean
Default	85.04	63.17	12.90	8.87	46.01	39.12	−47.07	−48.73
SOA/MS^-	85.34	63.62	12.88	8.80	46.05	38.90	−47.08	−48.59
G11	86.36	64.53	12.95	8.97	46.25	39.24	−47.18	−48.70
V10	86.39	64.81	13.02	9.04	46.28	39.34	−47.23	−48.85
G11-Internal	84.93	62.80	12.77	8.77	45.92	38.86	−47.07	−48.63
V10-Internal	84.84	64.81	12.92	8.88	46.15	39.34	−47.18	−48.66
Default-PI	56.83	47.96	8.46	6.64	41.75	36.05	−45.72	−47.48
Default-FN	113.51	90.83	17.81	13.51	51.16	45.10	−49.40	−51.57
G11-FN	114.68	92.27	17.95	13.62	51.37	45.26	−49.54	−51.72
G11-PI	58.11	49.68	8.60	6.84	42.03	36.47	−45.93	−47.72
G11-κ	86.11	64.82	12.93	8.94	46.09	39.06	−47.18	−48.72

[a] Averaged from ∼945–980 mb; [b] Calculated by using the grid-cell mean

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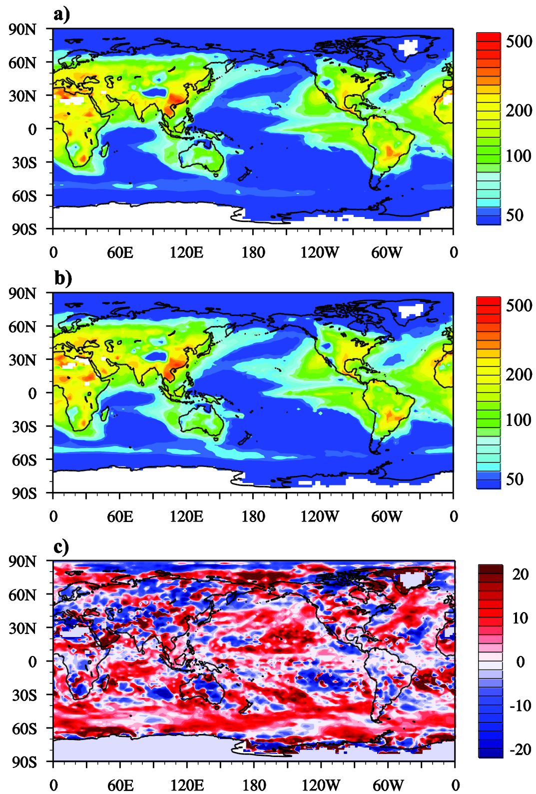

Fig. 1. Model predicted the 5-yr average in-cloud droplet number concentration (cm^{-3}) from 940–985 mb for the **(a)** Default and **(b)** G11 simulations and the **(c)** percentage change between the two simulations.

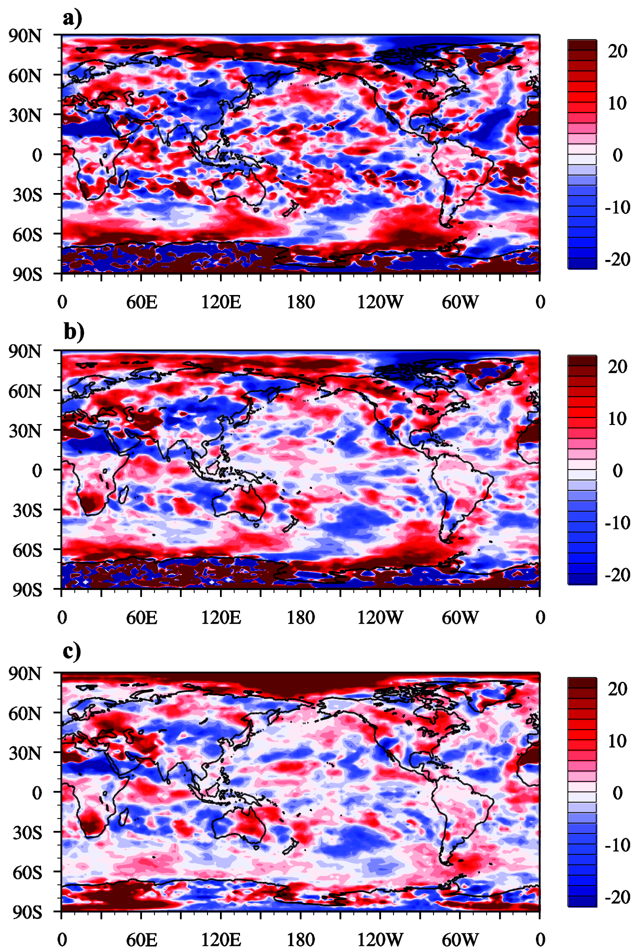

Fig. 2. Model predicted percentage change in the 5-yr average grid-mean **(a)** column cloud droplet number, **(b)** liquid water path, and **(c)** shortwave cloud forcing by marine emissions.

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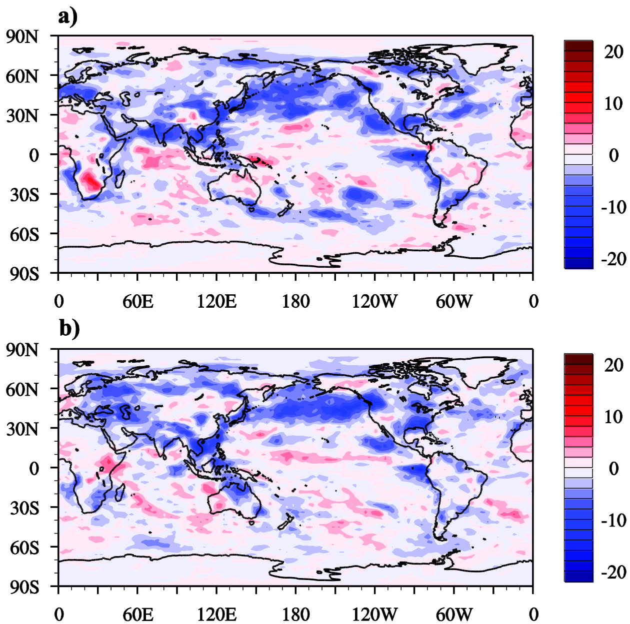

Fig. 3. Model predicted difference in the shortwave cloud forcing ($W\,m^{-2}$) between the **(a)** Default and Default-PI and between the **(b)** G11 and G11-PI simulations.

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