Effective radiative forcing in the aerosol—climate model CAM5.3-MARC-ARG [INCOMPLETE DRAFT; WORK IN PROGRESS]

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**Abstract.** We quantify the effective radiative forcing (ERF) of anthropogenic aerosols modelled by the aerosol—climate model CAM5.3-MARC-ARG. CAM5.3-MARC-ARG is a new configuration of the Community Atmosphere Model version 5.3 (CAM5.3) in which the default aerosol module has been replaced by the two-Moment, Multi-Modal, Mixing-state-resolving Aerosol model for Research of Climate (MARC). We compute differences between simulations using year-1850 aerosol emissions and year-2000 aerosol emissions in order to assess the ERF of anthropogenic aerosols. We compare the aerosol radiative effects produced CAM5.3-MARC-ARG with those produced by the standard configuration of CAM5.3, which uses the modal aerosol module with three log-normal modes (MAM3). Compared to MAM3, we find that MARC produces stronger cooling via the direct radiative effect, stronger cooling via the surface albedo radiative effect, and stronger warming via the cloud longwave radiative effect. Overall, MARC produces a global mean net ERF of W m-2, which is stronger than the global mean net ERF of W m-2 produced by MAM3. The geographical distribution of ERF also differs between MARC and MAM3. We conclude that the specific representation of aerosols in global climate models has important implications for climate modelling.

# 1 Introduction

Aerosol particles play an important role in the Earth’s climate system. There are three primary mechanisms by which aerosols can interact with radiation. First, aerosols interact directly with radiation by scattering and absorbing incoming sunlight (Haywood and Boucher, 2000). Second, aerosols interact indirectly with radiation via interactions with clouds, by acting as the cloud condensation nuclei on which cloud droplets condense and the ice nuclei that facilitate freezing of cloud droplets (Fan et al., 2016; Rosenfeld et al., 2014; Tao et al., 2012). Third, aerosols can influence the albedo of the Earth’s surface (Ghan, 2013).

The top-of-atmosphere radiative effect caused by anthropogenic emissions of aerosols and aerosol precursors is often referred to as the “effective radiative forcing” (ERF) of anthropogenic aerosols (Boucher et al., 2013). This is equivalent to “the radiative flux perturbation associated with a change from preindustrial to present-day [aerosol emissions], calculated in a global climate model using fixed sea surface temperature” (Haywood and Boucher, 2000). This approach “allows clouds to respond to the aerosol while surface temperature is prescribed” (Ghan, 2013).

The magnitude of the ERF of anthropogenic aerosols is highly uncertain. Estimates of the global mean total ERF of anthropogenic aerosols range from to W m-2 (Boucher et al., 2013). This large uncertainty in the aerosol ERF is a major source of uncertainty in projections of future climate change. Furthermore, the aerosol ERF is regionally inhomogeneous, contributing another source of uncertainty in climate projections (Shindell, 2014). The regional inhomogeneity of the aerosol ERF has likely also influenced rainfall patterns during the 20th century (Wang, 2015). In order to improve understanding of current and future climate, including rainfall patterns, it is necessary to improve understanding of the magnitude and regional distribution of the aerosol ERF.

The primary tools available for investigating the aerosol ERF are state-of-the-art global climate models. However, there is widespread disagreement among these models(Quaas et al., 2009; Shindell et al., 2013). Of particular importance are model parameterizations related to aerosol—cloud interactions, such as the activation scheme (Rothenberg et al., 2017), the choice of autoconversion threshold radius (Golaz et al., 2011), and constraints on the minimum cloud droplet number concentration (Hoose et al., 2009). The detailed representation of aerosols also likely plays an important role, because the aerosol and size distribution, chemical composition, and mixing-state determine hygrocopisity and hence influence activation (Kodros and Pierce, 2017; Petters and Kreidenweis, 2007).

In this manuscript, we investigate the uncertainty in aerosol ERF associated with the representation of aerosols. In particular, we assess the aerosol radiative effects produced by a new configuration of the Community Atmosphere Model version 5.3 (CAM5.3). In this new configuration of CAM5.3, the default modal aerosol module has been replaced by the two-Moment, Multi-Modal, Mixing-state-resolving Aerosol model for Research of Climate (MARC). We compare the aerosol radiative effects produced by this new configuration with those produced by the default modal aerosol module in CAM5.3.

# 2 Method

## 2.1 Modal aerosol modules (MAM3 and MAM7)

The Community Earth System Model version 1.2.2 (CESM 1.2.2) contains the Community Atmosphere Model version 5.3 (CAM5.3). Within CAM5.3, the default aerosol module is a modal aerosol module with three log-normal modes (MAM3) (Liu et al., 2012). Optionally, a more detailed model aerosol module with seven log-normal modes (MAM7) (Liu et al., 2012) can be used instead of MAM3. In this manuscript, we often refer to MAM3 and MAM7 collectively as “MAM”. The seven modes included in MAM7 are Aitken, accumulation, primary carbon, fine soil dust, fine sea salt, coarse soil dust, and course sea salt. Within each of these modes, MAM7 simulates the mass mixing ratios of internally-mixed sulphate, ammonium, primary organic carbon, secondary organic carbon, black carbon, soil dust, and sea salt (Liu et al., 2012). In MAM3, three simplifications are made: first, the primary carbon mode is merged into the accumulation mode; second, the fine soil dust and fine sea salt modes are also merged into the accumulation mode; third, the coarse soil dust and course sea salt modes are merged with each other; and fourth, ammonium is implicitly included via sulphate and is no longer explicitly simulated. As a result, MAM3 simulates just three modes: Aitken, accumulation, and coarse. This reduces the computational expense. More recently, a version containing four modes (MAM4) (Liu et al., 2016), has also been coupled to CAM5.3, but we do not consider MAM4 in this study.

The MAM aerosols interact with radiation, allowing aerosol direct and semi-direct effects to be represented. The aerosols can act as cloud condensation nuclei via the ARG activation scheme (Abdul-Razzak and Ghan, 2000) and can also act as ice nuclei. Via such activation, the aerosols are coupled to the stratiform cloud microphysics (Gettelman et al., 2010; Morrison and Gettelman, 2008), allowing aerosol indirect effects on stratiform clouds to be represented. These indirect effects dominate the effective radiative forcing due to aerosols in CAM version 5.1 (Ghan et al., 2012). In comparison to many other global climate models, this effective radiative forcing due to aerosols in CAM5.1 is relatively strong (Shindell et al., 2013).

## 2.2 The two-Moment, Multi-Modal, Mixing-state-resolving Aerosol model for Research of Climate (MARC)

The two-Moment, Multi-Modal, Mixing-state-resolving Aerosol model for Research of Climate (MARC) simulates the evolution of mixtures of aerosol species. MARC is based on the aerosol scheme by Wilson et al. (Wilson et al., 2001) and hence shares a common heritage with the HAM aerosol module used in the ECHAM5-HAM aerosol—climate model (Stier et al., 2005). Previous versions of MARC have used both in cloud-resolving model simulations (Ekman et al., 2004, 2006, 2007; Engström et al., 2008; Wang, 2005a, 2005b) and in global climate model simulations (Ekman et al., 2012; Kim et al., 2008, 2014).

As documented by Rothenberg et al. (Rothenberg et al., 2017), MARC has been coupled to the Community Atmosphere Model version 5.3 (CAM5.3) within the Community Earth System Model version 1.2.2 (CESM 1.2.2). In this configuration, MARC replaces the MAM3 aerosol module.

MARC tracks the number concentrations and mass concentrations of different lognormal modes. The externally-mixed modes include three pure sulphate modes (nucleation, Aitken, and accumulation), pure OC, and pure BC. The internally-mixed modes include mixed organic carbon plus sulphate (MOS) and mixed black carbon plus sulphate (MBS). In MOS, it is assumed that the organic carbon and sulphate are mixed homogeneously within each particle. In MBS, it is assumed that each particle contains a black carbon core surrounded by a sulphate shell. Mineral dust and sea-salt are represented using sectional single-moment schemes, each with four size bins. Further details can be found in Rothenberg et al. (Rothenberg et al., 2017). The Supplement of Rothenberg et al. (Rothenberg et al., 2017) also contains some validation of the aerosol fields simulated by MARC in comparison to observations.

In addition to interacting with radiation, the aerosols interact with the stratiform cloud microphysics scheme using standard stratiform cloud microphysics scheme, as would be the case with the standard MAM3 configuration of CAM5.3. Various aerosol activation schemes can be used with MARC (Rothenberg et al., 2017), including versions of a recently-developed polynomial chaos expansion scheme (Rothenberg and Wang, 2016, 2017). The choice of activation scheme can substantially influence the effective radiative forcing (Rothenberg et al., 2017). In order facilitate comparison between the MAM and MARC aerosol modules, we have chosen to keep the activation scheme constant in this study – hence, as is the case for the MAM simulations, the ARG activation scheme (Abdul-Razzak and Ghan, 2000) is also used for the MARC simulations. We refer to this configuration as “CAM5.3-MARC-ARG”.

## 2.3 Computational performance

In order to assess the computational performance of MARC, in comparison to MAM, six timing simulations have been performed. The configuration of these simulations is described in the caption of Table 1.

Before looking at the results, it is worth noting that the default radiation diagnostics differ between MARC and MAM. As highlighted by Ghan (Ghan, 2013), in order to calculate the direct radiative effect of aerosols, a second radiation call is required in order to diagnose “clean-sky” fluxes – in this diagnostic clean-sky radiation call, interactions between aerosols and radiation are switched off. In MARC, these clean-sky fluxes are diagnosed by default. However, in MAM, these clean-sky fluxes are not diagnosed by default, although simulations can be configured to include the necessary diagnostics. The inclusion of the clean-sky diagnostics increases computational expense. Hence, in order to facilitate a fair comparison between MARC and MAM, we have performed two simulations for each aerosol module: one with clean-sky diagnostics switched on, and one with clean-sky diagnostics switched off.

The results from the timing simulations are shown in Table 1. When clean-sky diagnostics are switched off, as would ordinarily be the case for long climate-scale simulations, MARC leads to only a 6% increase in computational cost compared to a standard configuration with MAM3. MAM7 is considerably more expensive. When clean-sky diagnostics are switched on, as is the case for the comparison simulations described below, the computational cost of MARC is very similar to that of MAM3.

## 2.4 Comparison simulations

In order compare results from MARC, MAM3, and MAM7, five simulations are performed:

1. “MAM3\_2000”, which uses MAM3 with year-2000 aerosol (and aerosol precursor) emissions;
2. “MAM7\_2000”, which uses MAM7 with year-2000 aerosol emissions;
3. “MARC\_2000”, which uses MARC with year-2000 aerosol emissions;
4. “MAM3\_1850”, which uses MAM3 with year-1850 aerosol emissions;
5. “MARC\_1850”, which uses MARC with year-1850 aerosol emissions.

The three simulations using year-2000 emissions, referred to as the “year-2000 simulations”, facilitate comparison of aerosol fields and cloud fields. The two simulations using year-1850 emissions, referred to as the “year-1850” simulations, further facilitate analysis of the radiative effects produced by MAM3 and MARC. The only difference between the year-2000 simulations and the year-1850 simulations is the aerosol (and aerosol precursor) emissions. In the figures and discussion of results, “2000-1850” refers to differences between the year-2000 simulation and the year-1850 simulation for a given aerosol module (e.g. MARC\_2000-MARC\_1850).

The emissions follow the default MAM emissions files, described in the Supplement of Liu et al. (Liu et al., 2012) and based on (Lamarque et al., 2010). 2.5% of the sulphur dioxide is emitted as sulphate. For the MAM simulations, the aerosol emissions from some sources follow a vertical profile (Liu et al., 2012). For the MARC simulations, sulphur emissions follow the same vertical profile; but all OC, BC and volatile organic compounds are emitted at the surface. Mineral dust and sea-salt emissions are not prescribed, being calculated “online”.

CESM 1.2.2, with CAM5.3, is used for all simulations. Greenhouse gas concentrations and sea-surface temperatures (SSTs) are prescribed using year-2000 climatological values, based on the “F\_2000\_CAM5” component set. The CAM5.3 atmosphere is run at a horizontal resolution of 1.9°×2.5° with 30 levels in the vertical. Clean-sky radiation diagnostics are included, facilitating diagnosis of the direct radiative effect. The Cloud Feedback Model Intercomparison Project (CFMIP) Observational Simulator Package (COSP) (Bodas-Salcedo et al., 2011) is switched on, although the COSP diagnostics are not analysed in this manuscript. Each simulation is run for 32 years, and the first one year and eleven months are excluded as spin-up – hence, an alaysis period of 30 years is available, with each analysis year starting in December.

## 2.5 Diagnosis of radiative effects

Pairs of prescribed-SST simulations, with differing aerosol emissions, facilitate diagnosis of aerosol effective radiative forcing via the “radiative flux perturbation” approach (Haywood et al., 2009). When clean-sky radiation diagnostics are available, the effective radiative forcing can be decomposed into contributions from different radiative effects (Ghan, 2013). Note: throughout this paper, we use the term “radiative forcing” only when referring to effective radiative forcing, defined as the radiative flux perturbation between a simulation using year-1850 emissions and a simulation using year-2000 emissions; we use the term “radiative effect” more generally.

The shortwave (SW) effective radiative forcing (*ERFSW*) can be decomposed as follows:

(1)

where ∆refers to the difference between the year-2000 and year-1850 simulations, *DRESW* is the direct radiative effect, *CRESW* is the clean-sky SW cloud radiative effect, and *SRESW* is the surface albedo radiative effect. These components are defined as follows:

(2)

(3)

(4)

(5)

where *F* is the net SW flux at top-of-atmosphere (TOA), *Fclean* is the clean-sky net SW flux at TOA, and *Fclean,clear* is the clean-sky clear-sky SW flux at TOA. It is worth noting that *Fclean,clear* is also sensitive to near-infrared absorption by water vapour – hence, *SRESW*, the so-called “surface albedo” radiative effect, can also be impacted by changes in water vapour.

The longwave (LW) effective radiative forcing (*ERFLW*) is calculated as follows:

(6)

where *L* is the net LW flux at TOA, *Lclear* is the clear-sky LW flux at TOA, and *CRELW* is the LW cloud radiative effect.

The net effective radiative forcing (*ERFSW+LW*) is simply

. (7)

All of the quantities mentioned Eqns. 1-7 are calculated at TOA. The analysis below will also consider absorption by aerosols in the atmosphere (*AAASW*), as follows:

(8)

where is the net SW flux at the Earth’s surface, and is the clean-sky net SW flux at the Earth’s surface.

# 3 Results

In this results section, we will focus on model output fields relating to different components of the effective radiative forcing, taking each component in turn: the direct radiative effect, the cloud radiative effect, and the surface albedo radiative effect. When discussing each of these components, we will also discuss related model fields – for example, in the section discussing the direct radiative effect we will also other fields related to aerosol—radiation interactions. But first, to provide context for the discussion of the radiative effects, we will examine the aerosol column burdens.

## 3.1 Aerosol column burdens

Aerosol column burdens, also referred to as aerosol column burdens, provide information about the total mass of a given aerosol species in an atmospheric column. Since the burdens are integrated throughout the atmospheric column, information about the vertical distribution of the aerosol is lost. However, the advantage of column burdens is that they are relatively simple to understand, facilitating comparison between the aerosol modules. However, before proceeding to examine to examine the column burdens, it is important to note that the representation of aerosol in MARC differs from that in MAM, meaning that the column burdens in MARC and MAM are not directly comparable, due to the different representations of mixing in MARC and MAM. In particular, MAM diagnoses column burdens for total sulphate, total OC, and total BC, whereas MARC diagnoses column burdens for pure OC, pure BC, MOS, MBS, and pure sulphate although total sulphate is also diagnosed. To facilitate comparison between MAM and MARC, we use the mass-mixing ratios diagnosed by MARC in order to calculate the total OC and total BC column burdens – the errors associated with this post-processing step are estimated to be less than 1% for all grid-boxes, and the errors are far smaller when global mean averaging is applied.

### 3.1.1 Total sulphate aerosol burden

Figure 1a—c shows the total sulphate aerosol burden results for the simulations using year-2000 emissions. For all three aerosol modules, the sulphate burden is highest in the Northern Hemisphere subtropics and mid-latitudes (Fig. 1a), especially near source regions with high anthropogenic emissions of sulphur dioxide (Fig. 1b, c). The sulphate burden is much lower in the Southern Hemisphere, especially over the remote Southern Ocean and Antarctica. There is generally close agreement between MAM and MARC over the Southern Hemisphere and the Northern Hemisphere tropics (Fig. 1a). However, over the Northern Hemisphere subtropics and mid-latitudes, MARC generally produces lower sulphate burdens than MAM3 does. Interestingly, the zonal mean sulphate burdens for MAM7 are very similar to those for MARC over the Northern Hemisphere sub-tropics (Fig. 1a).

When the simulations using year-1850 and year-2000 emissions are compared (Fig. 1d—f), it becomes evident that anthropogenic emissions of sulphur are responsible for more than half of the global mean sulphate burden. Both MAM3 and MARC produce widespread positive 2000-1850 differences across the Northern Hemisphere and also across South America, Africa, and Oceania.

### 3.1.2 Total organic carbon aerosol burden

Figure 2 shows results for the total OC aerosol burden. In contrast to the sulphate burden results discussed above, the year-2000 OC aerosol burden peaks in the tropics (Fig. 2a), especially sub-Saharan Africa and South America (Fig. 2b, c), due to emissions from wildfires. The impact of anthropogenic emissions of OC aerosol is evident over South Asia and East Asia (Fig. 2b, c). Generally, the OC aerosol burden produced by MARC is higher than that produced by MAM, with the MAM3 and MAM7 zonal mean burdens being very similar to one another (Fig. 2a).

MAM3 and MARC both produce positive 2000-1850 differences in OC aerosol burden over the major OC emissions regions, while MARC produces a stronger negative signal over North America than MAM3 does (Fig. 2d—f). These 2000-1850 differences arise due to changes in both wildfire emissions and anthropogenic emissions between year-1850 and year-2000.

### 3.1.3 Total black carbon aerosol burden

Figure 3 shows results for the total BC aerosol burden. For the year-2000 simulations, the BC aerosol burden is high over sub-Saharan Africa and South America (Fig. 3b, c), as was the case for the OC aerosol burden, due to large emissions of BC from wildfires. However, in contrast to the OC aerosol burden, the peak in zonal mean BC aerosol burden occurs in the Northern Hemisphere sub-tropics and mid-latitudes (Fig. 3a), due to anthropogenic emissions of BC over East Asia, South Asia, Europe, and North America. The results for MARC are generally similar to those for MAM in the tropics (Fig. 3a). Outside of the tropics, MARC generally produces a slightly highly BC aerosol burden than MAM3 does, especially over remote regions far away from sources (Fig. 3b, c), suggesting that the BC lifetime is longer in MARC than it is MAM3.

MAM3 and MARC produce similar increases in BC aerosol burden between year-1850 and year-2000 (Fig. 3d—f). In MARC, positive 2000-1850 differences are found over even remote ocean regions (Fig. 3f), consistent with a longer BC lifetime in MARC compared to MAM3.

## 3.2 Aerosol—radiation interactions and the direct radiative effect

### 3.2.1 Aerosol optical depth

Aerosols scatter and absorb shortwave radiation, leading to extinction of incoming sunlight. Before considering the direct radiative effect, we will first look at results for aerosol optical depth (AOD), a measure of the total extinction due to aerosols in an atmospheric column.

Figure 4 shows AOD results. In both MAM3 and MARC, emission of dust from deserts, especially the Sahara Desert, drive the highest annual mean AODs found globally (Fig. 4b, c), causing zonal mean AOD to peak in the Northern Hemisphere subtropics (Fig. 4a). In other regions, both anthropogenic aerosol emissions and natural aerosol emissions, including emissions of sea-salt, can dominate the AOD signal. The AODs produced by MARC are often much lower than those produced by MAM3, especially over subtropical ocean regions (Fig. a—c). The differences between the aerosol burdens in MAM3 and MARC, discussed above, are insufficient to explain the differences in the AODs. Hence it is likely that differences in the optical properties of the MARC aerosols and the MAM3 aerosols are responsible for the fact that MARC generally produces lower AODs. Differences in emissions of sea-salt may also play a role.

Positive 2000-1850 differences in aerosol burdens, discussed above, drive positive 2000-1850 differences in AOD (Fig. 4d—f). The 2000-1850 differences in AOD produced by MARC are much smaller than those produced by MAM3, consistent with the lower year-2000 AODs produced by MARC.

### 3.2.2 Direct radiative effect

Figure 5 shows direct radiative effect () results. reveals the influence of direct interactions between radiation and aerosols on the net shortwave flux at TOA (Eq. (3)). Aerosols that scatter efficiently, such as sulphate, generally contribute to negative values of , indicating a cooling effect at TOA. Aerosols that absorb SW radiation, such as BC, generally contribute to positive values of , indicating a warming effect at TOA. Other factors, such as the presence of clouds, the vertical distribution of aerosols relative to clouds, and the albedo of the Earth’s surface, also play a role in determining (Stier et al., 2007). Due to these factors, especially the differing impact of scattering and absorbing aerosols and variations in the albedo of the Earth’s surface, large AOD values may not necessarily correspond to large values of . Having said that, for the MAM3 and MARC simulations using year-2000 emissions, the spatial distributions of (Fig. 5b, c) share some similarities with those of AOD. Over dark ocean surfaces in the subtropics, scattering by aerosols drives negative values of . The impact of dust on differs between MAM3 and MARC, due to differing optical properties: in MAM3, absorption by dust drives positive values over the bright surface of the Sahara Desert, while little radiative impact is evident downwind over the dark surface of the tropical Atlantic Ocean; in MARC, scattering by dust drives negative values over the tropical Atlantic Ocean, while little radiative impact is evident over the Sahara Desert.

, the 2000-1850 difference in , produced by MAM3 is relatively weak at all latitudes (Fig. 5d, e), with a global mean of only -0.02±0.01 W m-2, due to the cooling effect of anthropogenic sulphur emissions being offset by the warming effect of increased emissions of BC aerosol (Ghan et al., 2012). In contrast, MARC produces a relatively strong cooling effect in the Northern Hemisphere (Fig. 5d), especially near anthropogenic sources of sulphur emissions (Fig. 5f), leading to a global mean of -0.18±0.01 W m-2.

### 3.2.3 Absorption by aerosols in the atmosphere

Figure 6 shows results for absorption of shortwave radiation by aerosols in the atmosphere (; Eq. (8)). Consideration of , which reveals heating of the atmosphere by aerosols, complements consideration of . For example, over the Sahara Desert, we noted above that the dust aerosol in MARC has little radiative impact at (Fig. 5c); however, Fig. 6c reveals that the dust aerosol in MARC leads to strong heating of the atmosphere. For both MAM and MARC, is largest near dust emission sources and BC emission sources (Fig. 6a—c). is generally weaker for MARC compared to MAM3.

, the 2000-1850 difference in , generally follows the same spatial distribution as the 2000-1850 difference in BC aerosol burden (Figs. 6d—f and 3d—f). Although dust dominated , dust has little impact on due to the dust emission in year-1850 being similar to that in year-2000. As with , is generally weaker for MARC compared to MAM3.

## 3.3 Aerosol—cloud interactions and the cloud radiative effects

### 3.3.1 Cloud condensation nuclei concentration

Many aerosol particles have the potential to become the cloud condensation nuclei (CCN) on which cloud droplets condense. Figure 7 shows the CCN concentration at a fixed supersaturation of 0.1% () in the bottom model level, near the Earth’s surface. Corresponding results, showing in the mid-troposphere, are shown in Figure S1 of the Supplement. Looking at for the year-2000 emissions (Figs. 7a—c and S1a—c), we make two initial observations. First, for both MAM and MARC, is generally higher in the Northern Hemisphere. Second, produced by MARC is generally much lower than that produced by MAM. When we look in more detail at the geographical distribution of produced by MAM3, and compare this to the column burden results, we notice that locations with high have either high or high . This suggests that both sulphate aerosol and OC aerosol act as efficient CCN in MAM, consistent with two previous MAM3-based studies that found that OC emissions from wildfires can exert a strong influence on clouds (Grandey et al., 2016a; Jiang et al., 2016). In contrast, the geographical distribution of produced by MARC closely resembles that of but does not resemble that of . This suggests that the OC aerosol produced by MARC is not an efficient source of CCN. This difference between MAM and MARC arises due to differences in the representation of OC aerosol, including mixing and hygroscopicity. Another difference between MAM and MARC is evident in the remote Southern Ocean 50—60°S: for MAM3, a local peak in suggests that sea-salt emission is providing a substantial supply of CCN; for MARC, no such peak is evident. However, it is worth noting that these results are for a fixed supersaturation of 0.1% -- as pointed out by Rothenberg et al. “*all* aerosol are potentially CCN, given an updraft sufficient enough in strength to drive a high-enough supersaturation such that they grow large enough to activate” (Rothenberg et al., 2017).

If we look at the results for , the 2000-1850 difference in (Figs. 7d—f and S1d—f), similar deductions about sulphate aerosol and OC aerosol can be made as were made above. For MAM3, the geographical distribution of reveals that changes in the availability of CCN are associated with changes in both and . For MARC, the geographical distribution of is associated with changes in , but is not closely associated with changes in . Both MAM and MARC generally produce positive values, revealing increasing availability of CCN between year-1850 and year-2000. The absolute increase is smaller in MARC than it is in MAM.

### 3.3.2 Column-integrated cloud droplet number concentration

The availability of CCN influences cloud microphysics via the formation of cloud droplets. Figure 8a—c shows column-integrated cloud droplet number concentration () for the year-2000 simulations. For MAM, is generally higher in the Northern Hemisphere, with very high values occurring over regions with abundant sulphate aerosol or OC aerosol providing abundant CCN. In contrast, for MARC there is no strong inter-hemispheric asymmetry in : there appears to be no influence from OC aerosol, consistent with the results discussed above; and the influence of sulphate aerosol appears weaker than in MAM. Interestingly, there is good agreement between MAM and MARC in the Southern Ocean – sea-salt appears to have a substantial influence on in MARC as well as in MAM, despite the fact the discrepancy in the results over the Southern Ocean.

When we look at , the 2000-1850 difference in (Fig. 8d—e), we see that anthropogenic emissions generally drive increases in , as expected. The absolute increase is smaller for MARC than for MAM, as expected.

### 3.3.3 Grid-box liquid and ice water paths

The availability of CCN also influences cloud water path. Figure 9a—c shows grid-box liquid water path () for the year-2000 simulations. is highest in the tropics and mid-latitudes. The geographical distribution of is similar to that of total cloud fractional coverage (Fig. S3a—c). The geographical distribution of produced by MARC is very similar to that produced by MAM. However, MARC produces slighted higher in the Southern Hemisphere mid-latitudes, the Southern Hemisphere subtropics, and the Arctic. It should be noted that relatively small changes in can have a large radiative impact.

Figure 10a—c shows grid-box ice water path () for the year-2000 simulations. As with , is highest in the tropics and mid-latitudes. The geographical distribution of is similar to that of high-level cloud fractional coverage (Fig. S6a—c). Although the geographical distribution of produced by MARC is very similar to that produced by MAM, MARC consistently produces lower values of than MAM does.

The 2000-1850 differences in and are shown in Figs. 9d—f and 10d—f. For MAM3, shows large increases over Europe, East Asia, Southeast Asia, South Asia, parts of Africa, and northern South America – the geographical distribution is similar to those of and for MAM3. For MARC, shows large increases over the same regions as for MAM3, with additional increases over Australia and North America. Overall, MARC produces larger than MAM3 does, especially over the Northern Hemisphere mid-latitudes. In comparison to MAM3, the relatively strong response produced by MARC contrasts with the relatively weak response and response produced by MARC.

Globally, the responses produced by MAM3 and MARC are relatively weak (Fig. 10d—f). However, relatively large values of , both positive and negative, are found regionally, and this regional response differs between MAM3 and MARC. In general, it appears that decreases in correspond to increases in and .

### 3.3.4 The clean-sky shortwave cloud radiative effect

Figure 11a—c shows the clean-sky shortwave cloud radiative effect (; Eq. (4)) for the year-2000 simulations. Clouds scatter much of the incoming solar radiation, exerting a strong cooling effect on the climate system. This cooling effect is strongest in the tropics and mid-latitudes. The geographical distribution is strongly negatively correlated with and (the total cloud water path; Fig. S2) – large values of and correspond to a strong cooling effect. The same applies to , the 2000-1850 difference in (Fig. 11d—f), which is strongly negatively correlated with and – increases in and drive a stronger shortwave cloud cooling effect. The cooling effect of is strongest in the Northern Hemisphere, particularly regions with high anthropogenic sulphur emissions, especially East Asia, Southeast Asia, and South Asia. Compared to MAM3, MARC produces a slightly stronger response in the mid-latitudes and a slightly weaker response in the sub-tropics. When globally averaged, both MAM3 and MARC produce a global mean response of approximately -2.1 W m-2.

### 3.3.5 The longwave cloud radiative effect

The cooling effect of is partially offset by the warming effect of (Eq. (6)), the longwave cloud radiative effect which arises due to absorption of longwave thermal infrared radiation. Figure 12a—c shows for the year-2000 simulations. As with the shortwave cooling effect, the longwave warming effect is strongest in the topics and mid-latitudes. The geographical distribution of is strongly positively correlated with and high-level cloud fraction (Fig. S6) – high ice cloud cover drives the longwave warming effect. The same is true for , the 2000-1850 difference in (Fig. 12d—f) – changes in high ice cloud cover drive changes in the longwave cloud warming effect. For both MAM3 and MARC, is positive over much of Southeast Asia, South Asia, the Indian Ocean, the Atlantic Ocean, and Pacific Ocean, while negative values of are found over much of Africa and parts of South America. When averaged globally, MAM3 produces a global mean of +0.54±0.02 W m-2, while MARC produces a stronger global mean of +0.66±0.02 W m-2. Hence offsets approximately one quarter of the cooling effect.

## 3.4 The surface albedo radiative effect

In addition to interacting with radiation both directly and indirectly via clouds, aerosols can influence the Earth’s radiative energy balance via changes to the surface albedo. Figure 13 shows , the 2000-1850 response of this surface albedo radiative effect (Eq. (5)). When globally averaged, is relatively small: W m-2 for MAM3, and a cooling effect of W m-2 for MARC. However, in the Arctic and high-latitude land regions of the Northern Hemisphere, can be relatively large. MAM3 produces a mixture of positive and negative values, averaging out to approximately zero globally. However, MARC tends to produce mainly negative values.

The response is associated with 2000-1850 changes in snow cover over both land and sea-ice (Fig. S7d—f) – increases in snow cover lead to negative values, while decreases in snow cover lead to positive values. It is unclear as to why the changes in aerosol emissions between year-1850 and year-2000 lead to these changes in snow cover. However, changes in snow rate (Fig. S8d—f) appear to be a major factor and can explain much of the snow cover response. Changes in black carbon deposition (Fig. S9d—f), leading to changes in the mass of black carbon in the top layer of snow (Fig. S10d—f), may also play a role. The mass of black carbon in the top layer of snow is much lower for MARC compared to MAM. The 2000-1850 difference in the mass of black carbon the top layer of snow is also much lower for MARC compared to MAM.

## 3.5 Net effective radiative forcing

The net effective radiative forcing (), which is the 2000-1850 difference in the net radiative flux at the top-of-atmosphere (Eq. (7)), is effectively the sum of the radiative effect components we discussed above (Eqs. (1) and(6)). Figure 14 shows . In general, the cloud shortwave component, , dominates, resulting in negative values of across much of the world. In particular, strongly negative values of , indicating a large cooling effect, are found near regions with substantial anthropogenic emissions of sulphur. The cooling effect is far stronger in the Northern Hemisphere than it is in the Southern Hemisphere. If coupled atmosphere—ocean simulations were to be performed, allowing sea-surface temperatures to respond, the large inter-hemispheric difference in effective radiative forcing would likely impact inter-hemispheric temperature gradients and hence impact rainfall patterns (Chiang and Friedman, 2012; Grandey et al., 2016b; Wang, 2015).

Across much of the world, the net cooling effect of produced by MARC is similar to that produced by MAM. However, in the mid-latitudes, MARC produces a stronger net cooling effect, especially over North America, Europe, and northern Asia. Another difference is that MARC appears to exert more widespread cooling over land than MAM does, while the opposite appears to be the case over ocean. These differences in the spatial distribution of may impact surface temperature gradients and rainfall patterns (see previous paragraph).

When averaged globally, MAM3 produces a global mean of W m-2. MARC produces a stronger global mean of W m-2. Considering that the produced by CAM5-MAM3 is strong compared to many other climate models (Shindell et al., 2013), the produced by CAM5-MARC-ARG is particularly strong.

# 4 Summary and conclusions

We have demonstrated that changing the aerosol module in CAM5.3 influences the radiative effects of the aerosols. Standard CAM5.3, which uses the MAM3 aerosol module, produces a global mean net ERF of W m-2 associated with the 2000-1850 difference in aerosol (and aerosol precursor) emissions. CAM5.3-MARC-ARG, which uses the MARC aerosol module, produces a stronger global net ERF of W m-2, a particularly strong cooling effect compared to other climate models (Shindell et al., 2013). The geographical distribution of ERF also differs between MAM3 and MARC.

By analysing the individual components of the net effective radiative forcing, we have demonstrated that:

1. The global mean 2000-1850 direct radiative effect produced by MAM3 ( W m-2) is close to zero due to the warming effect of black carbon aerosol opposing the cooling effect of sulphate aerosol and organic carbon aerosol. In contrast, the 2000-1850 direct radiative effect produced by MARC is W m-2, with the cooling effect of sulphate aerosol being larger than the warming effect of black carbon aerosol.
2. The global mean 2000-1850 shortwave cloud radiative effect produced by MARC ( W m-2) is very similar to that produced by MAM3 ( W m-2). However, the geographical distribution differs: for MAM3, the cooling peaks in the Northern Hemisphere subtropics; while for MARC, the cooling peaks in the Northern Hemisphere mid-latitudes.
3. The global mean 2000-1850 longwave cloud radiative effect, which partially offsets the shortwave cloud radiative effect, produced by MARC ( W m-2) is stronger than that produced by MAM3 ( W m-2).
4. The global mean 2000-1850 surface albedo radiative effect produced by MARC ( W m-2) is again stronger than that produced by MAM3 ( W m-2).

If climate simulations were to be performed using a coupled atmosphere-ocean configuration of CESM, these differences in the radiative effects produced by MAM3 and MARC would likely to differences in the climate response. In particular, the differences in the spatial distribution of the radiative effects would likely impact rainfall patterns (Wang, 2015). In light of these results, we conclude that the specific representation of aerosols in global climate models has important implications for climate modelling.

# Code and data availability

The MARC source code is available via <https://github.mit.edu/marc/marc_cesm/>. For this study, MARC commit “ff48dbe” has been used. Model namelist files, configuration scripts, and analysis code are available via <https://github.com/grandey/p17c-marc-comparison/> (doi:TODO). The model output data analysed in this paper are available via TODO (doi:TODO).

# Author contributions

AA and DR coupled MARC to CAM5.3 in CESM1.2.2, under the supervision of CW. AA, DR, QJ, and CW contributed to further development of CAM5.3-MARC-ARG, with DR being the primary software maintainer. HHL and BSG contributed to testing of CAM5.3-MARC-ARG. BSG and DR designed the experiment, with contributions from QJ and CW. BSG configured and performed the simulations. BSG, DR, and HHL analysed the results. BSG produced the figures shown in this manuscript. [TODO – statement about writing of manuscript.] CW provided supervisory guidance throughout the project.

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# Tables

Table 1: Results from six timing simulations. Each of these simulations consists of “20-day model runs with restarts and history turned off” (CESM Software Engineering Group, 2015), repeated five times in order to assess variability. The repetition of each simulation allows standard errors to be calculated via calculation of corrected sample standard deviation. For consistency, all runs have been submitted on the same day. For each run, 720 processors, spread across 20 nodes on Cheyenne (doi:10.5065/D6RX99HX), have been used. As with the year-2000 comparison simulations (Section 2.4), a model resolution of 1.9° × 2.5° is used, SSTs and greenhouse gas concentrations are prescribed using year-2000 climatological values, and aerosol and aerosol precursor emissions follow year-2000 emissions. In contrast to the comparison simulations, COSP has not been used in these timing simulations. The simulation costs shown represent the total cost of all model components, including components (such as the land scheme) that do not directly interact with the aerosol module.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Aerosol module | Clean-sky radiation diagnostics | Notes | CESM simulation cost  **±** standard error, (processor hours / model year) | Relative simulation cost (% above MAM3 with clean-sky diagnostics switched OFF) |
| MAM3 | OFF | Standard CAM5.3 | 325.5±0.8 | 0.0% |
| MAM7 | OFF | Standard CAM5.3 + MAM7 | 435.6±1.5 | +33.8% |
| MARC | OFF | Clean-sky diagnostics switched off via modification of source code | 344.3±0.7 | +5.8% |
| MAM3 | ON | Diagnostic clean-sky radiation call specified in simulation namelist | 362.3±0.8 | +11.3% |
| MAM7 | ON | Diagnostic clean-sky radiation call specified in simulation namelist | 472.2±1.0 | +45.1% |
| MARC | ON | Standard CAM5.3-MARC-ARG | 361.1±0.7 | +10.9% |

# Figures

[TO-DO: insert figures and write captions, to replace the links and text below.]

[Copies of the figures are available via [this link](https://drive.google.com/open?id=12ETElUb5i5xpWggwha4QdTBfQXjtLbEY).

The figures were produced using [figures\_draft\_2017b.ipynb](https://github.com/grandey/p17c-marc-comparison/blob/master/manuscript_draft2017b/figures_draft_2017b.ipynb).

For the zonal means, standard errors have been calculated using the annual zonal mean for each year and are indicated by shading. Often these standard errors are smaller than the width of the plotted lines, so the shading is not visible.

For the all maps, the area-weighted global mean and associated standard error, calculated using the annual global mean for each year, is shown below each map.

For the maps showing year-2000 results, white indicates values of zero unless otherwise specified (by a “±” value in the center of the colorbar).

For the maps showing 2000-1850 differences, white indicates differences with a magnitude less than the value indicated in the center of the colorbar. Stippling indicates locations where (i) the magnitude of the difference is greater than the value indicated in the center of the colorbar and (ii) the difference is statistically significant at a significance level of 0.05 after controlling the false discovery rate (Benjamini and Hochberg, 1995; Wilks, 2016). The two-tailed *p* values are generated by Welch’s unequal variances *t*-test, using annual mean data as the input. The approximate *p* value threshold, , which takes the false discovery rate into account, is written underneath each map.]