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Description and evaluation of a new 4-mode version of Modal Aerosol Module (MAM4) within version 5.3 of the Community Atmosphere Model

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4-Mode Version of Modal Aerosol Module in CAM5

X. Liu et al.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◀ | ▶

◀ | ▶

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

Atmospheric carbonaceous aerosols play an important role in the climate system by influencing the Earth's radiation budgets and modifying the cloud properties. Despite the importance, their representations in large-scale atmospheric models are still crude, which can influence model simulated burden, lifetime, physical, chemical and optical properties, and the climate forcing of carbonaceous aerosols. In this study, we improve the current 3-mode version of modal aerosol module (MAM3) in the Community Atmosphere Model version 5 (CAM5) by introducing an additional primary carbon mode to explicitly account for the microphysical ageing of primary carbonaceous aerosols in the atmosphere. Compared to MAM3, the 4-mode version of MAM (MAM4) significantly increases the column burdens of primary particulate organic matter (POM) and black carbon (BC) by up to 40 % in many remote regions, where in-cloud scavenging plays an important role in determining the aerosol concentrations. Differences in the column burdens for other types of aerosol (e.g., sulfate, secondary organic aerosols, mineral dust, sea salt) are less than 1 %. Evaluating the MAM4 simulation against in situ surface and aircraft observations, we find that MAM4 significantly improves the simulation of seasonal variation of BC concentrations in the polar regions, by increasing the BC concentrations in all seasons and particularly in cold seasons. However, it exacerbates the overestimation of modeled BC concentrations in the upper troposphere in the Pacific regions. The comparisons suggest that, to address the remaining model POM and BC biases, future improvements are required related to (1) in-cloud scavenging and vertical transport in convective clouds and (2) emissions of anthropogenic and biomass burning aerosols.

1 Introduction

Atmospheric aerosols from natural and anthropogenic sources play an important role in the climate system. In spite of extensive studies in the past several decades, radiative

forcing of atmospheric aerosols is still associated with large uncertainties according to the Intergovernmental Panel of Climate Change Assessment Reports (Forster et al., 2007; Myhre et al., 2013). The large uncertainty of aerosol radiative forcing among global climate models (GCMs) reflects the diversity in treatments of aerosol properties and processes in GCMs. A multitude of processes influence aerosols in the atmosphere, which include emission, nucleation, coagulation, gas-phase and aqueous chemical reactions, dry deposition, gravitational settling, and wet scavenging by clouds and precipitation. These processes determine the burden and lifetime of aerosols, particle size distribution, mixing state, and thus the radiative forcing of aerosols on the climate (Textor et al., 2006; Schulz et al., 2006; Quaas et al., 2009). Furthermore, the burden and size distribution of aerosols control their surface area density and thereby influence heterogeneous chemistry, which can then induce chemical effects such as extended methane lifetime (Tilmes et al., 2015).

Primary carbonaceous aerosols are produced from the incomplete combustion of fossil and biomass fuels, and are one of the most important types of aerosol in the atmosphere. Most models tend to speciate the mass of carbonaceous aerosols into a light-absorbing component (black carbon, BC) and a purely-scattering component (particulate organic matter, POM). BC has a strong warming effect, second only to carbon dioxide on the climate system, with the industrial-era (1750 to 2005) direct radiative forcing of $+0.71\,\mathrm{W\,m^{-2}}$ with 90 % uncertainty bounds of $(+0.08, +1.27)\,\mathrm{W\,m^{-2}}$ (Bond et al., 2013). Once emitted into the atmosphere, BC can be transported to the polar regions far from distant sources (e.g., Wang et al., 2013), and deposited on the surface of snow and sea ice by dry and wet deposition. BC-in-snow can absorb solar radiation efficiently, heat the snowpack, and induce a positive feedback on the surface albedo (Hansen and Nazarenko, 2004; Jacobson, 2004; Hansen et al., 2005; Flanner et al., 2007, 2009, 2012).

Although climatologically important, BC is still not simulated well by GCMs in many regions. For example, high bias of BC concentrations in the upper troposphere in the tropics and subtropics exists in almost all the global models participating in the Ae-

4-Mode Version of Modal Aerosol Module in CAM5

X. Liu et al.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◀ | ▶

◀ | ▶

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

roCom Phase I intercomparison (Koch et al., 2009; Schwarz et al., 2010), while low BC bias in the lower atmosphere by over a factor of 10 exists in the polar regions. The aerosol absorption optical depth also tends to be biased low in GCMs compared to satellite observations (Koch et al., 2009), although compared to ground-based sun photometers the models tend to be in better agreement (Kinne et al., 2006).

Freshly emitted carbonaceous aerosols are usually hydrophobic (i.e., water insoluble), especially those emitted from fossil fuel combustion. They cannot serve as cloud condensation nuclei (CCN), and are not able to nucleate cloud droplets. However, these particles can experience physical and chemical ageing in the atmosphere. Condensation and coagulation processes coat carbonaceous particles with soluble species (e.g., sulfate), which enables them to nucleate cloud droplets. Inside clouds, once these cloud droplets are converted to raindrops via various rain-production processes, carbonaceous aerosols in the cloud droplets can be removed from the atmosphere when raindrops fall to the ground (i.e., the so-called nucleation scavenging). Meanwhile, when BC is internally mixed with soluble materials, its absorption of sunlight can be enhanced by up to a factor of two (Jacobson, 2001, 2003). Therefore, in order to simulate aerosol concentration, spatial distribution, lifetime, and climate forcing correctly, a physically-based representation of atmospheric processes affecting the mixing state of carbonaceous aerosols is needed in the models.

The simplest treatment of carbonaceous aerosols, e.g., bulk models, assumes that carbonaceous aerosols and other aerosol types (e.g., sulfate) are externally mixed (i.e., different components do not coexist in the same particle), and often uses a prescribed time scale (1–2 days) for the ageing of primary carbonaceous aerosols from the hydrophobic to hydrophilic state (Cooke and Wilson, 1996; Tie et al., 2005). More sophisticated treatments, e.g., modal and sectional aerosol models, predict the aerosol size distribution, and determine the mixing state of carbonaceous aerosols by the coating thickness of soluble materials such as sulfate and organics on these aerosols (e.g., Liu et al., 2005; Stier et al., 2005; Spracklen et al., 2005; Bauer et al., 2008). Even more advanced aerosol treatments explicitly resolve the mixing state by tracking the

composition of individual particles in a population of different aerosol types (Riemer et al., 2009). However, this approach is computationally prohibitive for global models.

Liu et al. (2012) developed a modal aerosol module (MAM) for the Community Atmosphere Model version 5 (CAM5). The 7-mode version of MAM (MAM7) provides the benchmark simulation of aerosols in CAM5. In MAM7, the microphysical ageing of primary carbonaceous aerosols from the primary carbon mode to the accumulation mode through condensation and coagulation is explicitly treated, with a threshold coating thickness of three monolayers of sulfate. For the sake of computational efficiency, a simplified 3-mode version of MAM (MAM3) was developed for use as the default in CAM5. MAM3 is based on the MAM7 by merging the primary carbon mode with the accumulation mode, and assuming instantaneous internal mixing of primary carbonaceous aerosols with secondary aerosols (e.g., sulfate and secondary organic aerosols).

CAM5 with MAM3 is able to simulate many features of the observed spatial distribution of concentrations of different types of aerosol, aerosol optical depth, aerosol number and size distribution over different geographical regions of the world. However, similar to other global models (Koch et al., 2009), CAM5 significantly underestimates the near-surface BC concentrations in remote regions, e.g., in the Arctic (Liu et al., 2012; Wang et al., 2013; Ma et al., 2013a). While the underestimation of BC emission in Asia (e.g., Cohen and Wang, 2014) and in the high-latitudes of northern hemisphere (NH) (e.g., Stohl et al., 2013) can be an important factor, the excessively efficient scavenging of BC by liquid cloud processes (Wang et al., 2013) and the coarse horizontal resolution ($\sim$ 100–200 km) of the model (Ma et al., 2014) also contribute to this model bias.

To address this low BC bias found in CAM5 with MAM3 as well as in many GCMs, without significantly increasing the computational cost, we have developed a 4-mode version of MAM (MAM4) for the next generation of GCMs, including the version 6 of CAM (CAM6) and the US Department of Energy's Accelerated Climate Modeling for Energy (ACME).

In this paper, we provide a technical description and a first evaluation of MAM4. The paper is organized as follows. Section 2 describes MAM4. Section 3 introduces the sensitivity tests related to the microphysical ageing of primary carbonaceous aerosols and model horizontal resolution. Comparison of MAM4 with MAM3 and evaluation of MAM4 with observation data focusing on BC are given in Sect. 4. Conclusions are drawn in Sect. 5.

2   Model description

2.1   CAM5

CAM5 is the atmosphere component of the Community Earth System Model version 1 (CESM1.0) (Neale et al., 2012). Compared to its earlier versions, CAM5 is designed to simulate aerosol effects on stratiform clouds (i.e., indirect effect) through acting as CCN and ice nuclei (IN). MAM predicts the mass mixing ratios of internally mixed aerosol species within an aerosol mode and number concentrations of aerosol in that mode (Liu et al., 2012). A two-moment stratiform cloud microphysics scheme (Morrison and Gettelman, 2008; Gettelman et al., 2010) predicts the mass and number mixing ratios of cloud liquid and cloud ice, diagnoses the mass and number mixing ratios of rain and snow, and considers the complicated conversions among the cloud hydrometeors. The nucleation (i.e., in-cloud) scavenging of aerosols in stratiform clouds is treated consistently with the droplet activation in cloud microphysics (Abdul-Razzak and Ghan, 2000), by explicitly calculating aerosols in the cloud-liquid-borne state (Ghan and Easter, 2006). Aerosols can affect ice microphysics by homogeneous ice nucleation in cirrus clouds with temperatures lower than about −37 °C and heterogeneous ice nucleation in cirrus clouds and in mixed-phase clouds (Liu et al., 2007; Liu and Penner, 2005). However, aerosol scavenging by these ice nucleation processes is neglected in CAM5.

Other major components in CAM5 include (1) a stratiform cloud macrophysics scheme (Park et al., 2014) to estimate the cloudy volume (cloud fraction), (2) a moist

Printer-friendly Version

Interactive Discussion

turbulence scheme (Bretherton and Park, 2009) to explicitly simulate stratus-radiation-turbulence interactions, (3) a radiation scheme (Iacono et al., 2008) to more accurately treat the radiative effects of clouds and aerosol, (4) a shallow convection scheme (Park and Bretherton, 2009) to better simulate the observed spatial distribution of shallow convective activity, and (5) a modified deep convection scheme (Zhang and McFarlane, 1995) with the inclusion of sub-grid convective momentum transports (Richter and Rasch, 2008) and the updated closure (Neale et al., 2008).

2.2 MAM in CAM5

There are two versions of MAM available in CAM5: the default MAM3 and an optional MAM7 (Liu et al., 2012). MAM3 predicts the aerosol size distribution with three aerosol modes (Aitken, accumulation and coarse modes), while MAM7 includes seven aerosol modes (Aitken, accumulation, primary carbon, fine dust and fine sea salt, coarse dust and coarse sea salt modes). In MAM3 instantaneous ageing of primary carbonaceous particles is assumed by emitting them in the accumulation mode, while in MAM7 the ageing of the carbonaceous particles in the primary carbon mode due to the condensation of sulfuric acid (H_2SO_4 vapor), ammonia (NH_3), and the semi-volatile organics, and the coagulation of Aitken mode particles with primary-carbon mode particles are explicitly treated. Both sea salt and dust emissions are calculated on-line and are highly sensitive to the surface wind speed. Other processes in the atmosphere affecting aerosol properties (e.g., number/mass concentration, size, density, refractive index, chemical composition) include new particle formation, gas- and aqueous-phase chemistry, dry deposition and gravitational settling, water uptake, in-cloud (nucleation) and below-cloud scavenging, and release from evaporated cloud and rain drops. There are 15 transported aerosol tracers in MAM3 and 31 in MAM7.

2.3 Development of MAM4

In this study, we developed a 4-mode version of MAM (MAM4), which includes an additional primary carbon mode on top of MAM3 to explicitly treat the microphysical ageing of primary carbonaceous aerosols in the atmosphere. As in MAM7, fresh emit-
ted POM/BC particles are put in the newly added primary carbon mode. A criterion of 8 mono-layers of sulfate or equivalent amount of secondary organic aerosol (SOA) with the same increase in volume-weighted hygroscopicity as sulfate is required to convert POM/BC particles in the primary carbon mode to the accumulation mode. We use a value of 0 for the hygroscopicity (κ parameter) of POM, which is more repre-
sentative of POM produced from fossil fuel combustion. The hygroscopicity of POM from biomass burning source can be higher (0.06–0.30) (Liu and Wang, 2010). A non-zero hygroscopicity of POM allows in-cloud scavenging of POM/BC particles in the primary carbon mode before they are aged into the accumulation mode, and then the differences in the POM/BC concentrations between MAM3 and MAM7 are smaller (Liu
et al., 2012). A separate treatment of carbonaceous aerosols emitted from fossil fuel vs. biomass burning sources will be needed to seamlessly consider the different hygro-scopicity of POM. The κ value for BC is set to 0 to reflect its hydrophobic nature.

With the new primary carbon mode, three prognostic variables are added in MAM4 compared to MAM3: mass mixing ratios of POM and BC in the primary carbon mode
and number concentration of the primary carbon mode particles, which brings the total number of aerosol tracers from 15 in MAM3 to 18 in MAM4. The other aerosol trac-ers and precursor gases are the same as those in MAM3. The computational cost increases by $\sim 10\%$ for the standard-alone CAM5 with MAM4 compared to CAM5 with MAM3. As noted in Liu et al. (2012), CAM5 with MAM7 is 30% slower than CAM5 with
MAM3. Figure 1 shows the schematic of aerosol modes and associated aerosol tracers in MAM4.

3 Model configurations and experiments

Two sets of experiments were performed using version 5.3 of CAM (CAM5.3) with the new MAM4 (hereafter CAM5-MAM4), as listed in Table 1. The first set is to test the model sensitivity to the criterion of number of mono-layers required for the ageing of POM/BC particles from the primary carbon mode to the accumulation mode. We conducted four experiments with the number of mono-layers set to 1, 2, 4 and 8, respectively. A higher mono-layer criterion means that a larger sulfate or SOA coating thickness is needed to age the primary carbonaceous aerosols, and thus these particles will stay longer in the primary carbon mode before converting to the accumulation mode. As noted in Liu et al. (2012), for a non-hygroscopic particle with a 0.134 μm diameter, which is the volume-mean size for BC/POM emissions, the critical supersaturation in the case of 8 mono-layers of sulfate is 0.32 % based on the Köhler theory, in comparison to 0.49 % in the case of 3 mono-layers. These CAM5-MAM4 simulations were conducted for present-day (year 2000) emissions and climate conditions with freely evolving meteorological fields (e.g., winds and temperature) at $0.9° \times 1.25°$ (1° hereafter) horizontal resolution for 11 years. The last 10 year results are used for analysis. For comparison, an additional experiment using CAM5.3 with the default MAM3 was performed for 11 years at the same 1° horizontal resolution.

The second set of experiments is to test the model sensitivity to the horizontal resolution (see Table 1), since model resolution has been suggested to play an important role for the BC transport to the remote regions (Ma et al., 2013a, 2014). Four experiments were conducted at $1.9° \times 2.5°$ (2° hereafter), 1, 0.5, and 0.25° using the specified dynamics (SD) configuration where the model meteorology is strongly constrained by an external meteorological analysis (Ma et al., 2013a, b, 2015; Tilmes et al., 2015). To explore the behavior of MAM4 at higher horizontal resolution, we used the ECMWF Year of Tropical Convection (YOTC) high-resolution (0.15°) analysis to drive the model. The SD configuration has been recalibrated so that the model physics properly responds to the realistic meteorology (Ma et al., 2015). This approach facilitates the direct compari-

son between model simulations and field campaign measurements since the simulation of aerosol lifecycle (emission, transport, and deposition) is based on a realistic climate. These SD simulations were performed from 1 November 2008 to 1 January 2010, and the one-year results in 2009 were used for our analysis. The 8-monolayer criterion was used in this set of experiments. In addition, by comparing the second set of experiments with the SD configuration to the first set of experiments with the free-running configuration (e.g., MAM4L8 vs. MAM4R1), we can examine the effect of model meteorology on aerosol simulations. We also ran CAM5.3 with the default MAM3 at 1° resolution with the SD configuration for the comparison.

4 Results

4.1 Comparison of MAM4 with MAM3

Figure 2 shows the latitude and longitudinal distributions of annual mean column burdens of BC and POM from the set 1 experiments with CAM5-MAM4, in comparison with the default MAM3. BC and POM burdens have maxima in industrial regions (e.g., East Asia, Europe, and North America) and in biomass burning regions (e.g., Central and Southern Africa, South America, Indonesia, and Siberia), but a small fraction of BC and POM is transported to the Pacific and Atlantic Oceans, and to the Arctic. In general the Southern Hemisphere (SH) high-latitudes have the lowest BC and POM burdens. As expected, BC and POM burdens from CAM5-MAM4 simulations are much higher than those with MAM3 far from source regions. The increase in BC and POM burdens with MAM4 is most evident for the largest number of monolayers. This is more clearly seen in Fig. 3, which shows the relative (percentage) differences of annual mean burdens of BC and POM between MAM4 and MAM3 for the set 1 experiments. Compared to MAM3, the global total burden increase for MAM4L1 is 10 and 16 %, respectively for BC and POM, while it is 128 and 174 % for MAM4L8. The largest increase occurs over the remote oceans and polar regions where the background concentrations are

4-Mode Version of Modal Aerosol Module in CAM5

X. Liu et al.

low (see Fig. 2). An increase by up to a factor of 10 can be seen in the Arctic in the MAM4L8 case. In contrast, the increase is less than a factor of 2 (i.e., less than 100 %) in the regions over the continents where the major sources are. For a larger thresh-old number of mono-layers, carbonaceous aerosols stay longer in the primary carbon

5   mode, indicating a slower ageing. Aerosol particles in the primary carbon mode are not subject to wet removal, which allows more POM/BC to be transported to the remote regions. We note that non-carbonaceous aerosols (i.e., sulfate, SOA, mineral dust and sea salt) are affected very little (i.e., ∼ 1 % difference for dust, and less than 0.5 % difference for other aerosol species, which are about an order of magnitude smaller

10   than their interannual variabilities) by the introduction of the primary carbon mode. The number of CCN at 0.1 % supersaturation, which is important for the cloud formation and climate, changes by ∼ 6 % on the global mean.

Figure 4 shows the latitude and longitudinal distributions of annual mean BC and POM column burdens from the set 2 experiments. With an increase of the horizontal

15   resolution from 2 to 0.25° for CAM5-MAM4 simulations, BC and POM burdens both increase by 10–20 % on the global mean and by about a factor of 2 in the Antarctic. This increase is due to (1) the resolved meso-scale eddies at higher resolutions which leads to stronger poleward eddy transport of aerosol species, and (2) less frequent wet scavenging resulting from the reduced frequency of collocation between aerosols

20   and clouds at higher resolutions (Ma et al., 2014). Figure 5 shows the percentage differences of annual mean BC and POM burdens at 1, 0.5, and 0.25° relative to 2° for the set 2 experiments with CAM5-MAM4. With the increase of horizontal resolution, clearly more BC and POM are transported to the remote regions. The largest increase occurs in the Antarctic by up to a factor of 2. Other strong increases are seen in regions

25   away from major sources, such as the Arctic, western and eastern Pacific, East Asian and North American pollution outflow regions, indicating the enhanced transport out of the source regions at higher resolutions. This suggests that the sensitivity of POM/BC concentrations to resolution (Ma et al., 2015) can be an important factor for the aerosol-cloud interactions. The regions with the smallest relative increases are the POM/BC

source regions (e.g., East Asia, Southern Africa, South America), and the subtropical dry zones where cloud and precipitation scavenging of aerosols is relatively weak.

By comparing MAM4L8 (in Fig. 2) with MAM4R1 (in Fig. 4), all else being the same, CAM5 with the one-year specific dynamics gives $\sim 20\%$ lower BC and POM global burdens than the 10 year free-running simulation. This can be partly attributed to the inter-annual variability of aerosols in the MAM4L8 simulation. However, the primary cause of this difference is the different meteorological conditions between the SD and the free-running CAM5 that affect the transport and cloud processing of aerosols. In particular, we find that the free-running CAM5 produces a convectively less stable atmosphere, which enhances the vertical transport of aerosols to higher altitudes where the aerosol lifetimes are longer.

4.2 Annual global budgets of BC and POM

Tables 2 and 3 give the BC and POM budgets, respectively, for the sensitivity to the number of monolayers in the set 1 experiments. Most BC is removed from the atmosphere by wet deposition ($\sim 80\%$). However, with the increase in number of monolayers required for the microphysical ageing of BC from the primary carbon mode to the accumulation mode, the wet deposition term becomes somewhat less important and the dry deposition term more important. Most BC resides in the accumulation mode, indicating the relatively fast ageing of BC after it is emitted into the primary carbon mode. The ageing timescale of primary carbon mode BC (i.e., burden divided by the ageing flux from primary carbon mode to the accumulation mode) ranges from 0.24 days in MAM4L1 to 2.08 days in MAM4L8. With more monolayers and slower ageing of primary carbonaceous aerosols, more emitted BC remains in the primary carbon mode, and is not subject to cloud processing. For example, about 30 % of the total BC burden is in the primary carbon mode for the MAM4L8 experiment, compared to 6 % for the MAM4L1 experiment. The total BC burden increases from 0.087 (MAM4L1) to 0.123 Tg (MAM4L8), because the BC lifetime increases from 4.10 (MAM4L1) to 5.79 days (MAM4L8). Interestingly, BC burden in the accumulation mode does not change

much in these simulations, because the accumulation mode BC source (from ageing) and lifetime do not change much as the ageing criterion (monolayers) changes (Liu et al., 2012).

Similar results can be derived for POM (Table 3), except that POM emissions are ~ 6 times higher, and the burdens are 7–8 times higher than those of BC in these simulations. The longer lifetime of POM than that of BC reflects different spatial and temporal distributions of POM vs. BC emissions from different sources (e.g., fossil fuel and biomass burning) with respect to cloud and precipitation distributions. A higher percentage of POM is emitted from biomass burning emissions than for BC, and the biomass burning emissions tend to occur more in tropical dry seasons and boreal non-winter seasons. Also, it is assumed that POM/BC from biomass burning emissions is injected into higher altitudes (up to 6 km) and thus is subject less to wet removal by clouds, compared to the POM/BC from fossil fuel, which is emitted near the surface.

Tables 4 and 5 give the BC and POM budgets, respectively, for the set 2 experiments. BC and POM burdens increase by 12 and 7 %, respectively, from 2 to 0.25°, with the increase occurring in the accumulation mode. The nearly invariant POM and BC burdens in the primary carbon mode with resolution indicate that the microphysical ageing of primary carbonaceous aerosols is insensitive to the resolution. This is verified by the nearly constant ageing timescales of primary carbon mode POM and BC. The accumulation mode burden increases are due to longer lifetimes for the accumulation mode aerosol (e.g., 3.38 vs. 3.95 days for BC at 2 vs. 0.25° resolution). This is due to slower wet removal rates (i.e., wet deposition sinks divided by burdens) at higher resolution, even though the wet removal sinks are slightly greater at higher resolution, and the dry deposition sinks are correspondingly slightly lower. Specified dynamics (MAM4R1) reduces BC and POM burdens by 18 and 14 %, respectively, compared to free running (MAM4L8), with most of the decrease occurring in the accumulation mode.

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

4.3 Comparison with in situ observations

Next, we compare the simulations with a dataset obtained during the HIAPER (High-Performance Instrumented Airborne Platform for Environmental Research) Pole-to-Pole Observations (HIPPO) campaigns (HIPPO 1–5) over the remote Pacific from 80° N to 67° S in January and November 2009, March/April 2010, and June/July and August/September 2011 (Wofsy et al., 2011). Figure 6 shows the latitude-altitude cross section of the Single Particle Soot Photometer (SP2) measured BC concentrations during the HIPPO 1–5 campaigns, in comparison with CAM5-MAM4 simulated BC concentrations for the same time periods in the set 2 experiments. Here we only show model simulations from the set 2 experiments where meteorological fields from the YOTC analysis are used to drive the model simulations. The highest observed BC concentrations were in March/April 2010 in the NH mid-latitudes, resulting from strong transport of BC from the East Asian continents to the Pacific (Gao et al., 2014). Observed BC concentrations in the NH were lowest in August/September compared to other seasons, consistent with the strong wet scavenging of aerosols by the East Asian monsoon precipitation in the summer. Elevated BC concentrations in the NH high latitudes (e.g., Arctic) can be seen throughout the seasons, most evident in the spring time (March/April), with the maximum BC concentrations at 3–7 km, due to the transport from the NH mid-latitudes (Liu et al., 2011; Wang et al., 2013; Ma et al., 2013). BC concentrations in the SH over ~ 50° S had a maximum in August/September 2011, and were produced from South American and Australian biomass burning activities in the dry season. The poleward transport of BC in SH can also be seen at high altitudes over 5 km, particularly during August/September.

CAM5-MAM4 running at the four model resolutions is able to capture the maximum of BC concentrations in the NH mid-latitudes in March/April 2010. MAM4 shows a better agreement with the HIPPO measurements compared with MAM3 (Tilmes et al., 2015); however, the magnitude is still too low. This could be attributed to the underestimation of BC emissions in East Asia in the IPCC AR5 emission inventory used in

CAM5 (Lamarque et al., 2010), as reported by previous studies (e.g., Cohen and Wang, 2014). We also find that the simulated aerosol layers are at higher altitudes than the observations. The maximum of BC concentrations in MAM4 is generally between 5 and 10 km, suggesting that the model still has deficiency in the treatment of convective transport and scavenging of aerosols in CAM5 (Wang et al., 2013). This bias is also evident in model simulations in June/July and August/September 2011. Overall, the model overestimates BC concentrations in the upper troposphere. With the increase of model resolution more BC is transported to the polar regions, but increases occur primarily in the upper troposphere and lower stratosphere and is particularly noticeable in January 2009.

Figures 7 and 8 compare the modeled BC profiles from the various CAM5 simulations in set 1 and 2 experiments, respectively, with SP2 measured BC profiles during the HIPPO1 campaign over the Arctic and remote Pacific in January 2009 (Schwarz et al., 2010). Vertical profiles of observed mean BC concentrations show strong variability with latitude zones, and standard deviations of observed BC concentrations are also large. The observations show that BC concentrations increase with altitude in the SH high latitudes (60–67° S), indicating the poleward transport of BC in the upper troposphere by the general circulation. At mid-latitudes (20–60° N and 20–60° S), less vertical variation is observed. In the tropics (20° S–20° N), BC concentrations increase with altitude and reach a maximum near 700 hPa, likely from biomass burning, and then reduce rapidly with height. Over the NH high latitudes (60–80° N), a steady decreasing trend with height is found. All the model simulations are able to capture vertical distributions of BC at SH high latitudes as well as SH and NH mid-latitudes, although the MAM4 simulations with a larger number of mono-layers (e.g., MAM4L8) show too high BC concentrations at the SH high latitudes. The model simulations overestimate BC concentrations in the upper troposphere in all the latitude zones, while they significantly underestimate observed BC concentrations at lower altitudes (below 400 hPa) in the NH high-latitudes. The low bias of BC concentrations near the surface in the NH high latitudes is improved with MAM4, but there is still an obvious discrepancy

between the model simulations and observations. In addition, MAM4 predicts higher BC concentrations than those from MAM3 at all altitudes and all latitude zones in the set 1 experiments (Fig. 7). With the increase of number of monolayers required for the ageing of primary carbonaceous aerosols in MAM4, modeled BC concentrations increase by up to a factor of 10. The same is true for the higher BC concentrations with MAM4 than MAM3 for the set 2 experiments at most altitudes (Fig. 8). However, modeled BC concentrations from MAM3 at 1° are higher than those from MAM4 at 2° in the upper troposphere and lower stratosphere. Modeled BC concentrations increase substantially at these altitudes from 2 to 1°. However, the changes are much smaller from 1 to 0.25° at all altitudes for all latitude zones.

Figures 9 and 10 compare observed BC vertical profiles from four aircraft campaigns in the tropics (CR-AVE and TC4), NH subtropics (AVE-Houston) and NH mid-latitudes (CARB) with simulations from the set 1 and set 2 experiments, respectively. Observed BC mean concentrations show a strong reduction from the boundary layer to the free troposphere in the tropics (CR-AVE and TC4) and in the subtropics (AVE-Houston), while there is less vertical variation of BC concentrations in the upper troposphere. For the AVE-Houston, CR-AVE, and TC4 campaigns, the model experiments with different numbers of monolayers required for ageing of primary carbonaceous aerosols over-estimate BC concentrations in the mid- and upper troposphere (Fig. 9), but compare better with observations in the lower troposphere. The CARB campaign in the mid-latitudes of North America in June encountered biomass burning plumes, and elevated BC concentrations were observed at ∼ 800 hPa, which are not captured by model simulations using the AR5 emission inventory. The SD simulations at higher resolutions in the set 2 experiments (Fig. 10) do not improve the overall agreement with observations, which indicates the necessity of improving the model physics (e.g., aerosol processing in convective clouds) and aerosol emissions (e.g., for biomass burning aerosols). The modeled BC concentrations do not change significantly with different model resolutions, although there is an increasing trend with increasing the resolution.

Figures 11 and 12 compare observed BC vertical profiles from two aircraft campaigns in the NH high latitudes (ATCTAS and ARCPAC) with the set 1 and set 2 experiments, respectively. Observed BC concentrations show less vertical variations from the surface to ~ 300 hPa than those in the tropics and NH mid-latitudes, especially in the spring. This results from the poleward transport of BC from NH mid-latitudes at high altitudes. Model simulations in the set 1 experiments underestimate observed BC concentrations below altitudes of ~ 300–400 hPa by 1–2 orders of magnitude, especially in spring (Fig. 11). Introducing the primary carbon mode with the number of monolayers increasing from 1 to 8 substantially increases the modeled BC concentrations in the spring, in much better agreement with observations. However, the improvement of BC simulation in the summer is less substantial. The difference of BC concentrations between MAM3 and MAM4 and among different MAM4 experiments is much smaller in the summer, similar in magnitude to the changes in the tropics and NH mid-latitudes (Fig. 9). Model simulations in the set 2 experiments show the increase of BC concentrations with MAM4, compared to MAM3, but the increase of model resolution does not produce a significant increase of BC concentrations at altitudes below ~ 200 hPa (Fig. 12).

Figures 13 and 14 show the seasonal variations of observed BC surface concentrations at four polar sites, in comparison with the model results from MAM4 set 1 and 2 experiments, respectively, along with the results from MAM3. Observed BC concentrations at the three Arctic sites show a strong seasonality with higher concentrations in boreal spring and winter, and much lower concentrations in boreal summer (by about one order of magnitude). Observed BC concentrations at the Antarctic site are lower by one order of magnitude than those in the Arctic, and the seasonality is weaker and different, with highest BC concentrations in the austral spring and summer. At the Arctic sites, CAM5 with MAM3 substantially underestimates the observed BC concentrations, especially in winter by more than two orders of magnitude, and produces an opposite seasonality to the observed one. This indicates the too efficient wet scavenging of BC during its transport from the mid-latitudes, especially in spring and winter, since

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

uncertainty of the emissions cannot explain these large discrepancies. CAM5-MAM4, especially with the larger number of monolayers significantly increases the BC concentrations at the Arctic sites, especially in winter, and thus improves the modeled seasonality of BC concentrations. Simulated BC concentrations also increase at the Antarctic site, but the seasonality is still wrong. Increasing the model resolution of CAM5-MAM4 results in a general increase of BC concentrations in different seasons, especially in summer at the Arctic sites, which however, weakens the simulated seasonality.

5 Conclusions

To reduce the biases of carbonaceous aerosol simulated in CAM5, a new 4-mode version of MAM (MAM4) has been developed by adding a primary carbon mode to the default 3-mode version (MAM3), and by explicitly treating the microphysical ageing of primary carbonaceous aerosols from the primary carbon mode to the accumulation mode. The computational time increases slightly by 10 %, comparing to CAM5 with MAM3. As expected, CAM5-MAM4 increases BC and POM burdens and enhances BC transport to remote regions (e.g., Arctic) due to the reduced wet scavenging of BC with more BC residing in the primary carbon mode. The increases are stronger with increasing number of hygroscopic monolayers required for the ageing of primary carbonaceous aerosols. The global burdens of BC and POM increase from 0.087 to 0.123 Tg and from 0.63 to 0.96 Tg, respectively, as the number of ageing monolayers is increased from 1 to 8. The BC and POM burdens increase by a factor of 2–10 in the polar regions, depending on the number of monolayers. The global mean lifetimes of BC and POM increase from 4.10 to 5.79 days and from 4.62 to 7.02 days, respectively.

We also explored the sensitivity of modeled carbonaceous aerosol to the horizontal resolution of CAM5-MAM4, as previous studies suggested its importance for the BC transport to remote regions (Ma et al., 2014). The global burdens of BC and POM increase from 0.098 to 0.110 Tg and from 0.81 to 0.87 Tg, respectively, as the resolution increases from 2 to 0.25°. The lifetimes of BC and POM increase from 4.61 to 5.18 days

and from 5.91 to 6.35 days, respectively. The largest relative increases in BC and POM burdens occur in the Antarctic, the Arctic, the western and eastern Pacific, and in the East Asian and North American pollution outflows. The increased resolution enhances the transport of aerosols by better resolving mesoscale eddies and the separation of

5 aerosol plumes from clouds and precipitation (Ma et al., 2014).

Modeled BC was compared to observations obtained from aircraft campaigns and at polar surface sites. For the vertical profiles of BC concentrations, CAM5-MAM4 increases the BC concentrations at all altitudes compared to MAM3. Therefore, MAM4 improves the modeled BC in the lower troposphere, especially in the Arctic. The mod-

10 eled near-surface BC concentrations at the Arctic sites have stronger increases in boreal winter and spring than in summer, which improves the modeled seasonal variation of BC concentrations. However, modeled near-surface BC concentrations in the polar regions are still lower by one order of magnitude compared to observations, even with the number of monolayers of 8 and with the model resolution of 0.25°. This points to

15 the potential problems of emissions and/or other model processes besides the ageing of primary carbonaceous aerosols discussed in this study. Wang et al. (2013) found that the wet scavenging of BC might be too efficient in CAM5 due to high liquid cloud fraction in the NH high latitudes, when comparing the wet scavenging rate with that from the multi-scale modeling framework (MMF). The MMF, which embeds a cloud-

20 resolving model in each climate model grid column, gives a much better simulation of BC concentrations in the Arctic (Wang et al., 2011). In addition, the aerosol emission inventories used in our simulations also likely contribute to the BC low biases in the Arctic. They neglect gas flaring emissions in and around the Arctic, and they may underestimate forest fire emissions in Siberia and Alaska as well as fossil fuel emissions

25 in East Asia and other developing countries.

While the modeled BC low bias in the lower troposphere is significantly reduced in the Arctic, the overestimation of BC concentrations in the mid- and upper troposphere over the Pacific and in the tropics and subtropics of North America is exacerbated with MAM4, based on the comparisons with several aircraft campaigns. With a unified

treatment of vertical transport and in-cloud wet removal in convective clouds in CAM5, Wang et al. (2013) significantly reduced the biases of aerosol distributions in the remote free troposphere predicted by CAM5. An ongoing joint effort between PNNL and NCAR is underway to merge the MAM4 with this unified treatment of convective transport and scavenging. This emphasizes the importance of improving the representations of clouds and precipitation and aerosol-cloud interactions in GCMs.

We note that in this study, we only present the results and analysis of primary carbonaceous aerosols, especially BC, since there have been more BC measurements, and POM is co-emitted with BC. The lifecycle and global budgets of other aerosol species such as sulfate, SOA, sea salt and dust are not presented in this study, since introducing the primary carbon mode has a negligible effect on these aerosol species. This is expected since sulfate and SOA primarily reside in the accumulation mode, while dust and sea salt are mostly in the accumulation and coarse modes. Changes in modeled CCN concentrations are also small. The change in the aerosol "effective" indirect forcing (i.e., radiative flux perturbation at the top of the atmosphere due to aerosol changes between the preindustrial and present-day times) induced by the MAM4 implementation is less than $0.1\,\mathrm{W\,m^{-2}}$ in the global mean.

Code availability

The release of CESM version 1.2.0 (with CAM5.3) can be obtained at (http://www.cesm.ucar.edu/models/cesm1.2/). Code modifications for the 4-mode version of Modal Aerosol Module are available upon request by contacting the corresponding author.

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◄ | ►►

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Discussion Paper

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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Table 1. Model experiments. Set 1 experiments are to test the model sensitivity to the criterion of number of ageing monolayers, running CAM5-MAM4 in free-running simulations at horizontal resolution of 1°. Set 2 experiments are to test the model sensitivity to the model horizontal resolution, running CAM5-MAM4 in specified dynamics (SD) simulations with the number of ageing monolayer of 8. In both sets of experiments the standard CAM5.3 with MAM3 is running for the comparison.

	Experiment Name	Mono-layer	Horizontal Resolution	Simulation Type
Set 1	MAM4L1	1	1°	Free-running
	MAM4L2	2	1°	Free-running
	MAM4L4	4	1°	Free-running
	MAM4L8	8	1°	Free-running
	MAM3	N/A	1°	Free-running
Set 2	MAM4R2	8	2°	SD
	MAM4R1	8	1°	SD
	MAM4R0.5	8	0.5°	SD
	MAM4R0.25	8	0.25°	SD
	MAM3R1	N/A	1°	SD

Table 2. Global BC budgets for monolayer sensitivity simulations in set 1 experiments.

	MAM3	MAM4L1	MAM4L2	MAM4L4	MAM4L8
Sources ($Tg\,yr^{-1}$)	7.76	7.76	7.76	7.76	7.76
Emission	7.76	7.76	7.76	7.76	7.76
Sinks ($Tg\,yr^{-1}$)	7.75	7.75	7.75	7.75	7.74
Dry deposition	1.30	1.38	1.42	1.50	1.64
Wet deposition	6.45	6.37	6.33	6.25	6.10
Lifetime (days)	3.95	4.10	4.30	4.80	5.79
Burden (Tg)	0.084	0.087	0.091	0.102	0.123
in accumulation	0.084	0.082	0.082	0.083	0.083
in primary carbon	0.000	0.005	0.009	0.019	0.040
Ageing timescale of					
primary carbon (days)	N/A	0.24	0.44	0.96	2.08

Table 3. Same as Table 2, except for POM budgets.

	MAM3	MAM4L1	MAM4L2	MAM4L4	MAM4L8
Sources ($Tg\,yr^{-1}$)	50.2	50.2	50.2	50.2	50.2
Emission	50.2	50.2	50.2	50.2	50.2
Sinks ($Tg\,yr^{-1}$)	50.1	50.1	50.1	50.1	50.1
Dry deposition	7.5	8.1	8.4	9.0	10.0
Wet deposition	42.6	42.0	41.7	41.1	40.1
Lifetime (days)	4.34	4.62	4.96	5.69	7.02
Burden (Tg)	0.60	0.63	0.68	0.78	0.96
in accumulation	0.60	0.59	0.58	0.59	0.59
in primary carbon	0.00	0.049	0.097	0.19	0.37
Ageing timescale of					
primary carbon (days)	N/A	0.37	0.74	1.48	2.97

Table 4. Global BC budgets for resolution sensitivity simulations in set 2 experiments.

	MAM3R1	MAM4R2	MAM4R1	MAM4R0.5	MAM4R0.25
Sources ($Tg\,yr^{-1}$)	7.76	7.76	7.76	7.76	7.76
Emission	7.76	7.76	7.76	7.76	7.76
Sinks ($Tg\,yr^{-1}$)	7.75	7.75	7.75	7.75	7.74
Dry deposition	1.57	2.10	2.01	1.90	1.77
Wet deposition	6.18	5.65	5.74	5.85	5.97
Lifetime (days)	3.25	4.61	4.77	5.02	5.18
Burden (Tg)	0.069	0.098	0.101	0.107	0.110
in accumulation	0.069	0.064	0.068	0.071	0.076
in primary carbon	0.000	0.034	0.033	0.036	0.034
Ageing timescale of primary carbon (days)	N/A	1.80	1.73	1.88	1.77

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

◀ | ▶|

◀ | ▶

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Table 5. Same as Table 4, except for POM budgets.

	MAM3R1	MAM4R2	MAM4R1	MAM4R0.5	MAM4R0.25
Sources ($Tg\,yr^{-1}$)	50.2	50.2	50.2	50.2	50.2
Emission	50.2	50.2	50.2	50.2	50.2
Sinks ($Tg\,yr^{-1}$)	50.1	50.1	50.2	50.1	50.1
Dry deposition	8.5	12.0	11.5	11.1	10.4
Wet deposition	41.6	38.1	38.6	39.1	39.7
Lifetime (days)	3.64	5.91	6.03	6.27	6.35
Burden (Tg)	0.50	0.81	0.83	0.86	0.87
in accumulation	0.50	0.48	0.50	0.52	0.55
in primary carbon	0.00	0.33	0.33	0.34	0.32
Ageing timescale of primary carbon (days)	N/A	2.68	2.66	2.73	2.56
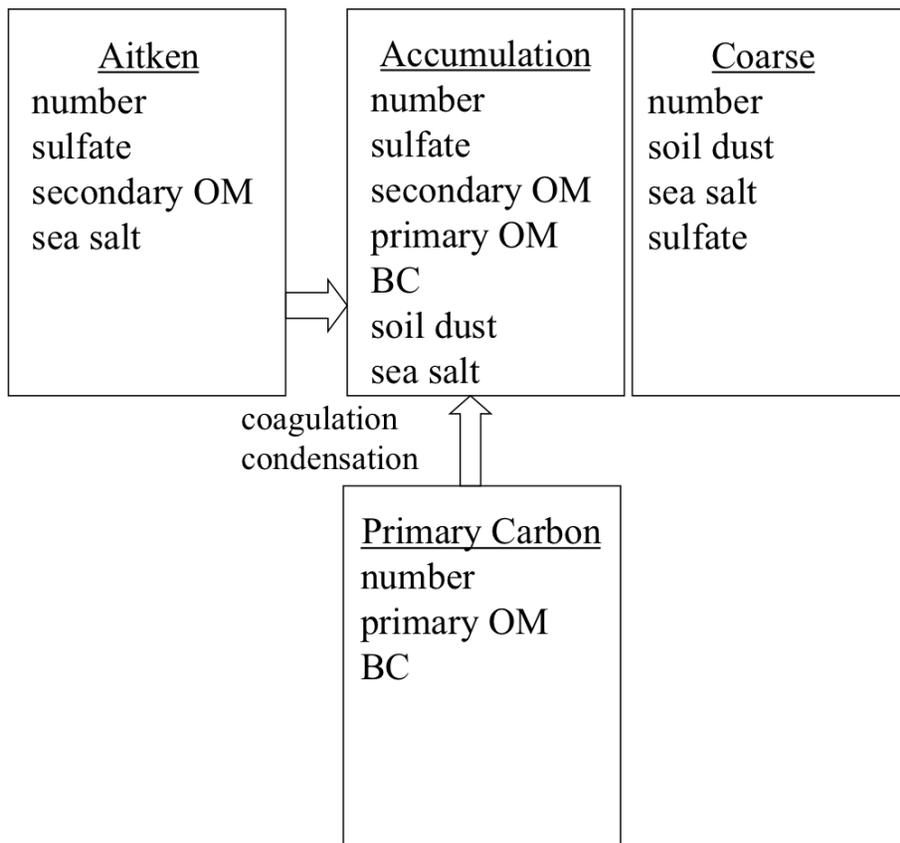
Figure 1. Schematic of aerosol modes and associated aerosol tracers in MAM4.
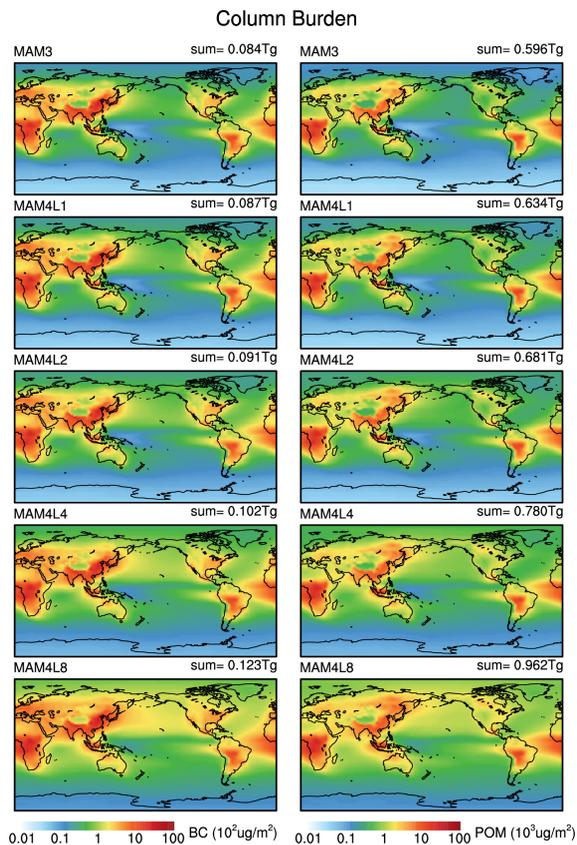
Figure 2. Latitude and longitudinal distributions of annual mean column burdens of BC (in $10^2\,\mu g\,m^{-2}$, left) and POM ($10^3\,\mu g\,m^{-2}$, right) from the set 1 experiments with the default MAM3, and MAM4 with different criteria of number of monolayers (i.e., MAM4L1, MAM4L2, MAM4L4, and MAM4L8).

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper |

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
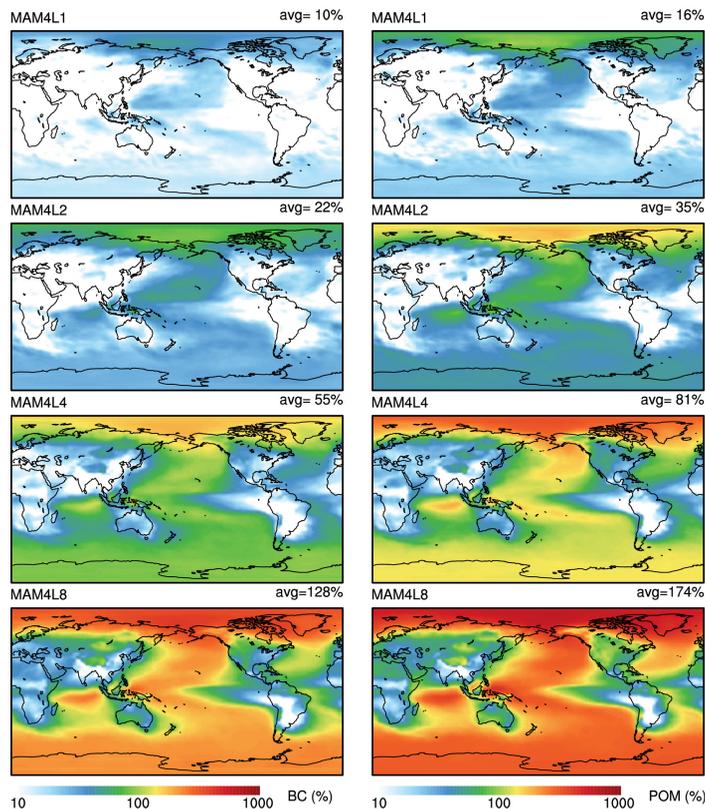
Figure 3. Latitude and longitudinal distributions of the relative (percentage) differences of annual mean burdens of BC (left) and POM (right) between MAM4 with different criteria of number of monolayers (i.e., MAM4L1, MAM4L2, MAM4L4, and MAM4L8) and MAM3 in the set 1 experiments.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
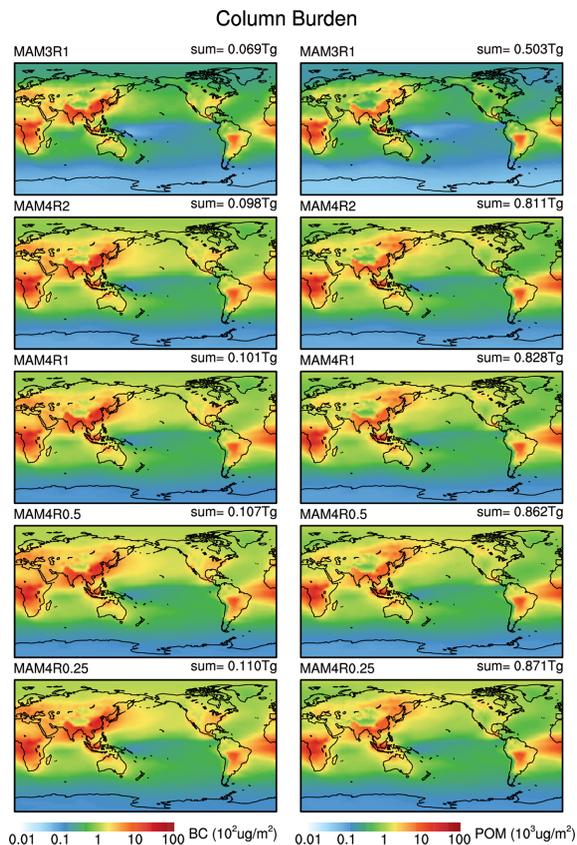
Figure 4. Latitude and longitudinal distributions of annual mean column burdens of BC (in $10^2 \, \mu g \, m^{-2}$, left) and POM ($10^3 \, \mu g \, m^{-2}$, right) from the set 2 experiments with the default MAM3 at $1°$ horizontal resolution (MAM3R1), and MAM4 at different horizontal resolutions (i.e., MAM4R2, MAM4R1, MAM4R0.5, and MAM4R0.25).

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
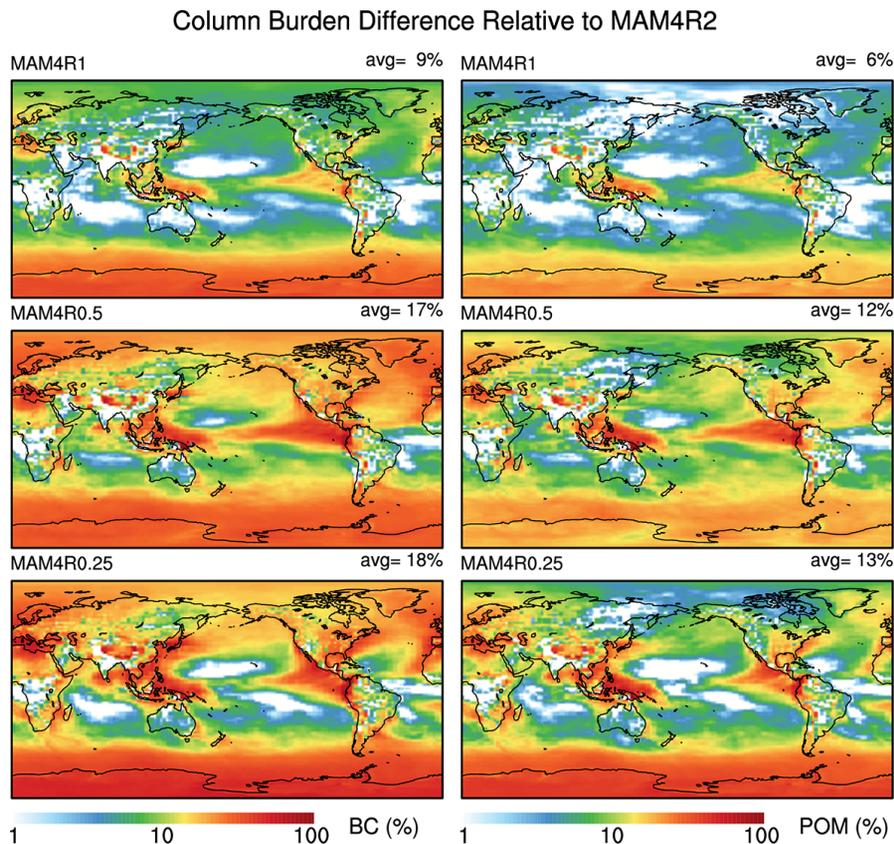
Figure 5. Latitude and longitudinal distributions of the relative (percentage) differences of annual mean burdens of BC (left) and POM (right) from the set 2 experiments with MAM4 at 1, 0.5, and 0.25° relative to 2° horizontal resolution.
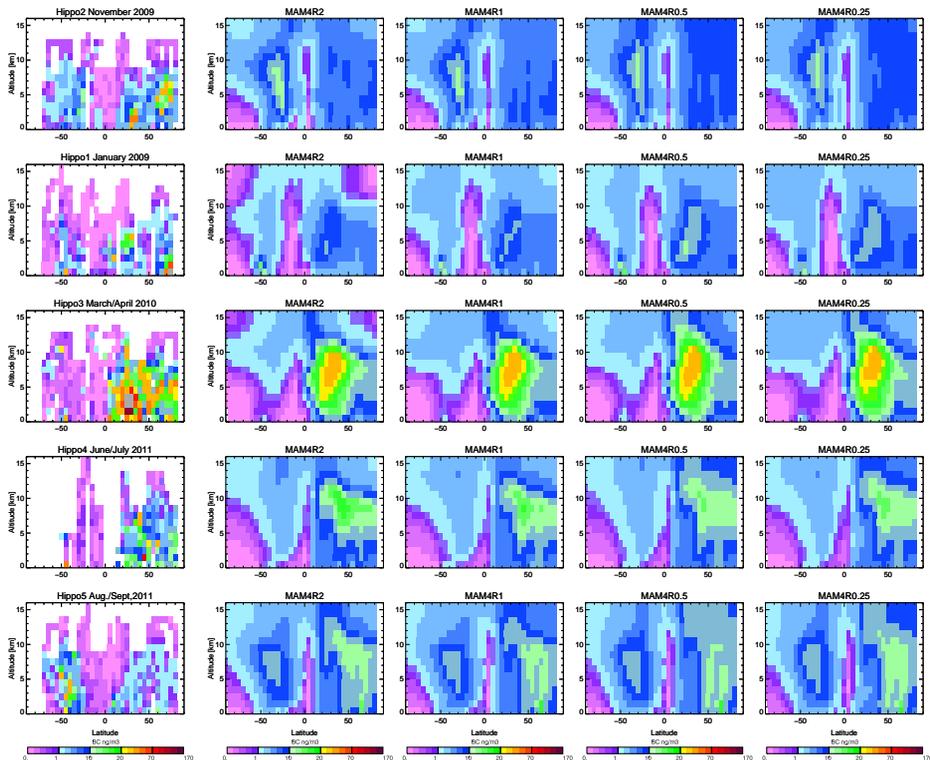
Figure 6. Latitude and altitude cross section of observed and simulated BC concentrations (in $ng\,m^{-3}$) during the HIAPER (High-Performance Instrumented Airborne Platform for Environmental Research) Pole-to-Pole Observations (HIPPO) campaigns (HIPPO 1–5) over the remote Pacific from 80° N to 67° S in January and November 2009, March/April 2010, and June/July and August/September 2011. MAM4 simulated BC concentrations for the same time periods in the set 2 experiments at different horizontal resolutions (i.e., MAM4R2, MAM4R1, MAM4R0.5, and MAM4R0.25) are used for comparison.
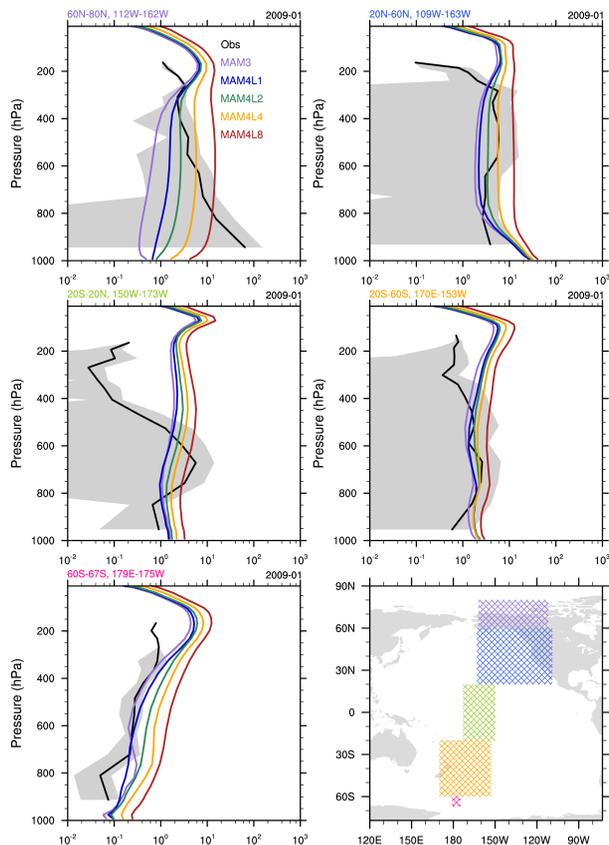
Figure 7. Observed and simulated vertical profiles of BC concentrations (in ng kg^{-1}) during the HIPPO1 campaign over the Arctic and remote Pacific in January 2009. The CAM5 simulations in the set 1 experiments (i.e, MAM3, MAM4L1, MAM4L2, MAM4L4, and MAM4L8) are used for comparison.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion
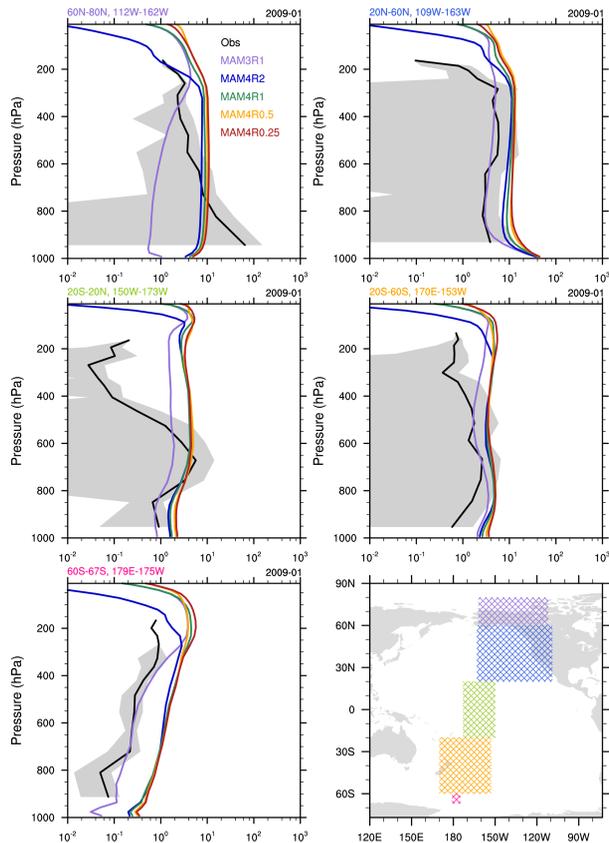
Figure 8. Same as Fig. 7, except for CAM5 simulations in the set 2 experiments (i.e., MAM3R1, MAM4R2, MAM4R1, MAM4R0.5, and MAM4R0.25).

Figure 9. Observed and simulated vertical profiles of BC concentrations (in $ng\,kg^{-1}$) from four aircraft campaigns in the tropics, NH subtropics and NH mid-latitudes: AVE-Houston (NASA Houston Aura Validation Experiment) in November 2010 and 2012, respectively, CR-AVE (NASA Costa Rica Aura Validation Experiment) in February 2006, TC4 (Tropical Composition, Cloud and Cl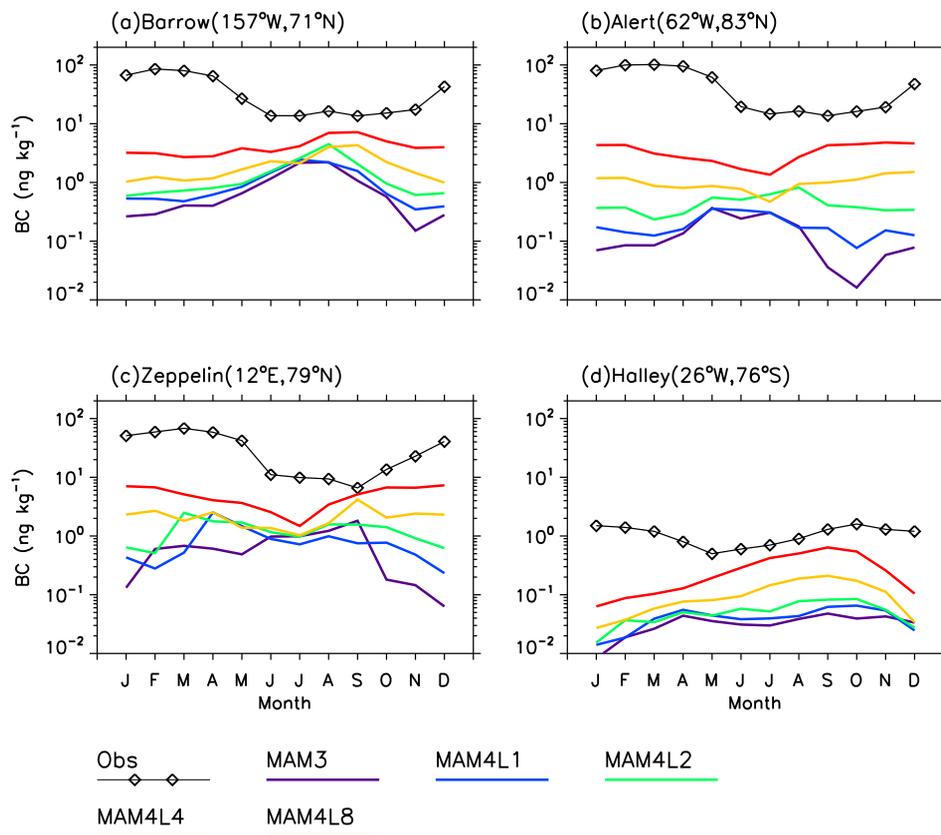imate Coupling) in August 2007, and CARB (NASA initiative in collaboration with California Air Resources Board) in June 2008. Observations are averages for the respective campaigns with dark solid curves for observation means, dark dashed curves for observation medians, and shaded areas for the plus/minus one standard deviation of observations. Simulated profiles for the CAM5 simulations in the set 1 experiments (i.e., MAM3, MAM4L1, MAM4L2, MAM4L4, and MAM4L8) are averaged over the points on the map and the indicated month.

Figure 10. Same as Fig. 9, except for CAM5 simulations in the set 2 experiments (i.e., MAM3R1, MAM4R2, MAM4R1, MAM4R0.5, and MAM4R0.25).
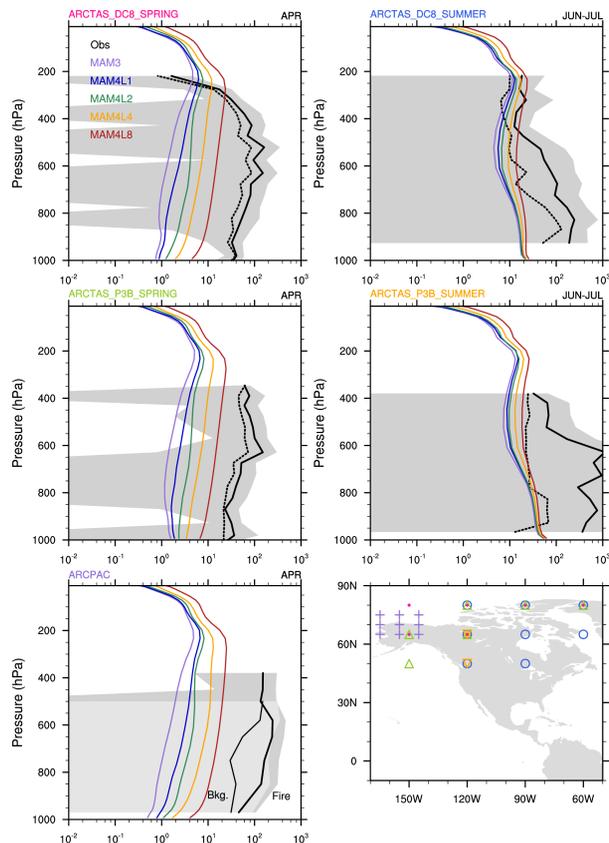
Figure 11. Same as Fig. 9, except for BC profiles at NH high latitudes from two other aircraft campaigns: ARCTAS (NASA Arctic Research of the Composition of the Troposphere from Aircraft and Satellite) in spring (April) and summer (June–July) 2008, and ARCPAC (NOAA Aerosol, Radiation, and Cloud Processes affecting Arctic Climate) in spring (April) 2008.
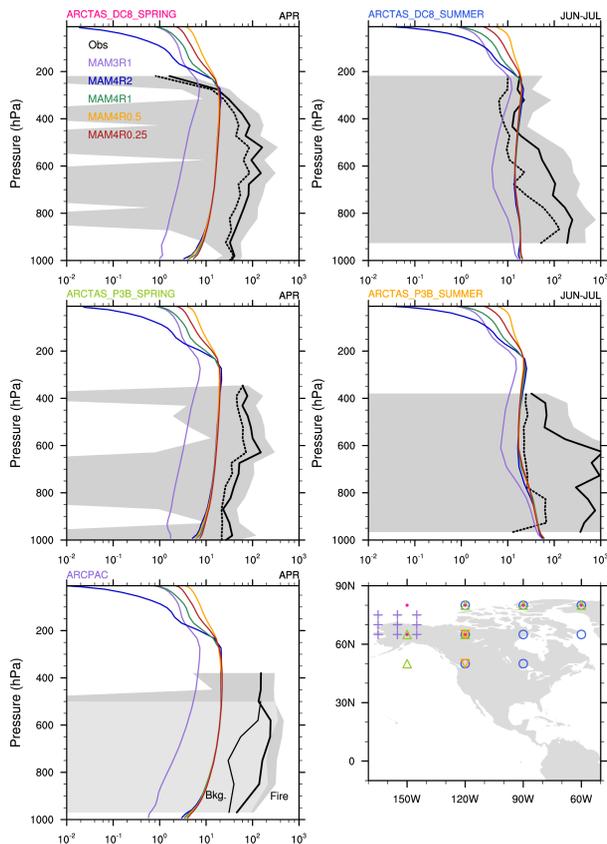
Figure 12. Same as Fig. 11, except for CAM5 simulations in the set 2 experiments.

Figure 13. Seasonal variations of observed and simulated BC surface concentrations (in ng kg^{-1}) at four polar sites. CAM5 simulations in the set 1 experiments (i.e., MAM3, MAM4L1, MAM4L2, MAM4L4, and MAM4L8) are used for comparison.

Title Page

Abstract | Introduction

Conclusions | References

Tables | Figures

|◄ | ►|

◄ | ►

Back | Close

Full Screen / Esc

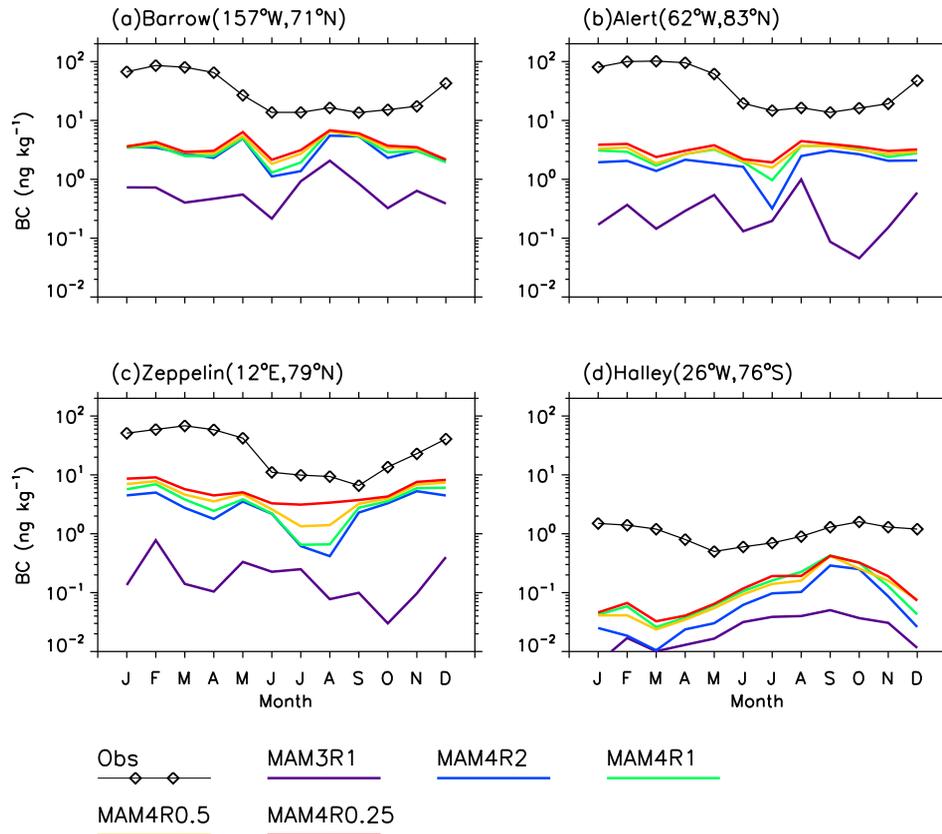Printer-friendly Version

Interactive Discussion

Figure 14. Same as Fig. 13, except for CAM5 simulations in the set 2 experiments (i.e., MAM3R1, MAM4R2, MAM4R1, MAM4R0.5, and MAM4R0.25).