Black Carbon Absorption Efficiency Under Preindustrial and Present-Day Conditions Simulated by a Size- and Mixing-State-Resolved Global Aerosol Model

Hitoshi Matsui

1Graduate School of Environmental Studies, Nagoya University, Nagoya, Japan

Abstract The positive radiative forcing of black carbon (BC) aerosol depends not only on the spatial and temporal distribution of BC but also on its absorption efficiency. The mass absorption cross section (MAC) of BC is enhanced by atmospheric aging processes that increase particle size and non-BC coating amounts (mixing state) of BC-containing particles. However, the representation of MAC (or absorption enhancement) in current global aerosol models has a large uncertainty. This study used a global aerosol model that resolves particle size and mixing state to show that the MAC of anthropogenic BC has increased by 50% from preindustrial to present-day conditions (from 5.6 to 8.6 m² g⁻¹) because faster present-day aging processes increase the fraction of thickly coated BC particles, which have high absorption efficiency. This effect is apparent only when the model considers BC mixing state with sufficient resolution. The impact of this MAC enhancement on BC direct radiative forcing is estimated to be 0.051–0.086 W m⁻² globally (22–37% of anthropogenic BC direct radiative forcing, 0.23 W m⁻²) and exceeds 0.5 W m⁻² near-source regions in East Asia. Sensitivity simulations show that BC direct radiative forcing and MAC are highly sensitive to non-BC emissions, secondary aerosol formation, and aerosol size distribution and mixing state in emissions. We must therefore improve our understanding of these factors by further observations and reduce their discrepancies between models to achieve better estimates of BC absorption efficiency and radiative forcing.

1. Introduction

Black carbon (BC) aerosol is emitted from the combustion of fossil fuels, biofuels, and biomass and plays an important role in Earth’s radiative balance through the absorption of solar radiation, both in the atmosphere (e.g., Bond et al., 2013; Chung & Seinfeld, 2005; Jacobson, 2000; Ramanathan & Carmichael, 2008) and on snow and ice surfaces (e.g., Clarke & Noone, 1985; Flanner et al., 2007; Hansen & Nazarenko, 2004) and on Earth’s ongoing climate change (Boucher et al., 2013; Myhre, Shindell, et al., 2013). Understanding of BC properties relevant to climate, such as the amounts of emissions, spatial and temporal distributions, microphysical and optical properties, and radiative effects, has rapidly advanced in the last decade, but their estimation still has large uncertainties (e.g., Bond et al., 2013; Samset et al., 2018). For example, the ranges of BC direct radiative forcing (DRF) since the Industrial Revolution were estimated to be 0.08–1.27 W m⁻² by Bond et al. (2013) (90% uncertainty bounds from 1750 to 2005 for all sources) and 0.05–0.37 W m⁻² in the intercomparison of global aerosol models of Myhre, Samset, et al. (2013) (the maximum/minimum range of 15 models from 1850 to 2000 for fossil fuel and biofuel sources). Reducing the uncertainties in the climatic impacts of BC is crucial for understanding the importance of aerosols in Earth’s ongoing climate change (Boucher et al., 2013; Myhre, Shindell, et al., 2013). The representation of BC microphysical and optical properties in models is a cause of the large uncertainties in estimations of the climatic impacts of BC. Condensation, coagulation, and oxidation processes in the atmosphere (called aging processes) gradually coat BC particles with non-BC species such as sulfate, nitrate, and organic aerosols (OA) and enhance two key properties: their hygroscopicity and their absorption efficiency (Moteki et al., 2007; Shiraiwa et al., 2008). The enhancement of hygroscopicity, which changes BC-containing particles from hydrophobic to hydrophilic and promotes their activity as cloud...
condensation nuclei, is an important factor for modeling the spatial distribution and lifetime of BC (e.g., He et al., 2016; Matsui, 2016b; Riemer et al., 2010; Stier et al., 2006; X. Liu et al., 2016). The enhancement of absorption efficiency leads to higher absorption aerosol optical depth (AAOD) and DRF (e.g., Fierce et al., 2016; Jacobson, 2001; Matsui, 2016a; Peng et al., 2016), although enhanced hygroscopicity, which reduces the BC burden by efficient wet removal, may offset some of this increase (Boucher et al., 2016; Stier et al., 2006). The mass absorption cross section (MAC) of BC, defined as BC absorption cross section per unit BC mass, is a key parameter for estimating AAOD and DRF. Previous studies have suggested that MAC is sensitive to BC mixing state, being around 5 m² g⁻¹ for fresh and uncoated (externally mixed) BC particles and >10 m² g⁻¹ for aged and coated (internally mixed) BC particles (Bond & Bergstrom, 2006; Boucher et al., 2016; Ram & Sarin, 2009; Zanatta et al., 2016).

The particle size and mixing state of BC are key microphysical properties for accurately estimating the hygroscopicity, MAC, absorption enhancement, and radiative effect of BC-containing particles. I and coworkers have recently developed a global aerosol model that resolves particle size and mixing state by using a two-dimensional sectional representation with 12 particle size bins (from 1 nm to 10 μm in dry diameter) and 8 mixing state bins (pure BC, six divisions of internally mixed BC, and BC-free particles) (Matsui, 2017; Matsui & Mahowald, 2017). Using this model, we have shown the advantages of size- and mixing-state-resolved simulations for understanding the global distribution, long-range transport, absorption, and direct radiative effect (DRE) of BC (Matsui & Mahowald, 2017; Matsui, Hamilton, & Mahowald, 2018; Matsui, et al., 2018).

This paper shows, by estimates of BC absorption efficiency in preindustrial and present-day conditions, that absorption efficiency has been enhanced substantially (by 50% for anthropogenic BC) as aging speed has historically increased. This enhancement can be simulated only when BC mixing state is resolved sufficiently in our global aerosol model. This paper also estimates BC DRF from preindustrial to present-day conditions and evaluates the impact of the enhanced absorption efficiency on BC DRF estimates. Finally, this paper shows the importance of non-BC emissions, secondary aerosol formation, and uncertainties in emissions for estimates of BC absorption efficiency and DRF.

2. Model Simulations

2.1. CAM5-chem/ATRAS Model

This study used the Community Atmosphere Model Version 5 (CAM5) (Lamarque et al., 2012; X. Liu et al., 2012; Neale et al., 2012) with the Aerosol Two-dimensional bin module for foRmation and Aging Simulation (ATRAS) (Matsui, 2017; Matsui, Koike, Kondo, Fast, et al., 2014). The CAM5-chem/ATRAS model (Matsui, 2017; Matsui & Mahowald, 2017) considers emissions, gas-phase chemistry (Emmons et al., 2010), nucleation (Kulmala et al., 2006; Matsui et al., 2011; Vehkamäki et al., 2002), condensation of sulfate, nitrate, and secondary OA (SOA) (Matsui, 2017; Matsui, et al., 2014), coagulation (Jacobson et al., 1994; Matsui, 2017; Matsui et al., 2013), cloud activation (Abdul-Razzak & Ghan, 2000, 2002), aqueous-phase chemistry (Tie et al., 2001), dry and wet deposition (Rasch et al., 2000; Wesely, 1989; X. Liu et al., 2012; Zender et al., 2003), and aerosol-radiation and aerosol-cloud interactions (Gettelman et al., 2010; Iacono et al., 2000; Morrison & Gettelman, 2008). SOA formation, which contributes to BC aging processes, is calculated based on the volatility basis set approach (Matsui, 2017; Matsui, et al., 2014).

The multiple mixing state representation used in this study has 12 particle size bins to represent particles from 1 to 10,000 nm in dry diameter and eight mixing state bins for fine particles (40–1,250 nm diameter) to classify pure BC, thinly coated BC, moderately coated BC, thickly coated BC, and BC-free particles. Mixing state bins have BC mass fractions of 0.0–0.0001, 0.0001–0.1, 0.1–0.2, 0.2–0.5, 0.5–0.8, 0.8–0.9, 0.9–0.99, and 0.99–1.0. Optical properties are specified in look-up tables based on the Mie theory with the shell-core (BC core and non-BC shell) treatment for internally mixed BC particles and the homogeneously mixed treatment for pure BC and BC-free particles (Matsui, 2017). Because this representation calculates size/mixing-state-dependent hygroscopicity and optical properties for individual two-dimensional bins, it can explicitly calculate the enhancement of absorption (without using a constant absorption enhancement factor) and hygroscopicity (without using a predetermined threshold between hydrophobic and hydrophilic BC) by aging processes.
The single mixing state representation, used here for comparison, resolves particle size using the 12 bins but assumes that all of the particles in each bin have the same chemical composition (internally mixed BC). This representation cannot properly consider changes in mixing state by aging processes and the resulting impacts on the lifetime and radiative effect of BC. Matsui, Hamilton and Mahowald (2018) showed that the BC radiative effect is overestimated when the single mixing state representation is used and that the resolution of BC-free, thinly coated BC, and thickly coated BC is important in models.

### 2.2. Simulation Setups

This study mainly used simulations with emissions in 1750 (preindustrial) and 2010 (present-day) from the Coupled Model Intercomparison Project Phase 6 (CMIP6) for aerosols and their precursor species (Hoesly et al., 2018; van Marle et al., 2017). CMIP5 emissions, which are available between 1850 (preindustrial) and 2000 (present-day) (Lamarque et al., 2010), were used for comparison and are listed in the supporting information. Both data sets yield the same main conclusions with respect to BC absorption efficiency (supporting information Figures 1 and 2) even though their time spans are not the same. Simulations for a future condition (the Year 2100) were performed using the Representative Concentration Pathway 4.5 (RCP4.5) emissions (Moss et al., 2010) and are discussed in section 3.3. Online simulations were performed for 6 years for each emission data set (preindustrial, present-day, and future) and mixing state representation (multiple or single), and the averages of the last 5 years were used for analysis. The model has a horizontal resolution of $1.9^\circ \times 2.5^\circ$ and has 30 vertical layers from the surface to ~40 km altitude. All simulations in this study relied on present-day climatological data for the ocean and ice surface boundary and long-lived climate forcers. Meteorological fields were not forced or nudged by any data. Because aerosol-cloud-radiation interactions were considered explicitly, meteorological fields are not the same in the different simulations.

The size distribution and mixing state of anthropogenic and biomass burning aerosol emissions are the same as in the Base-size case of Matsui, Hamilton, and Mahowald (2018): Number median diameters are 70 nm for fossil fuel combustion sources and 100 nm for biofuel combustion and biomass burning sources, with a geometric standard deviation of 1.8 for all sources. These values are generally consistent with previous observational studies for urban atmospheres (e.g., Kondo et al., 2010) and biomass burning plumes (e.g., Kondo et al., 2011). Emitted particles were assigned either a pure BC or BC-free mixing state. Although some or many BC particles are known to be internally mixed near emission sources (Cheng et al., 2009; China et al., 2013), the mixing state distributions of emitted BC particles (in terms of eight mixing state bins) are not known well and are highly uncertain. They are especially uncertain in the preindustrial time because there are no direct observations of the mixing state distributions of emitted BC particles in the preindustrial time. Considering that point, this study assumed this simple mixing state to investigate the maximum impact of the mixing-state-resolved treatment. I examined the importance of the mixing state of emitted particles through sensitivity simulations, described in section 3.4, assuming that half of BC is emitted as internally mixed particles composed of BC and OA.

### 2.3. Evaluations by Observations

The CAM5-chem/ATRAS model has been evaluated previously for its reproduction of aerosol mass and number concentrations and optical properties by using various surface and aircraft measurements (Matsui & Mahowald, 2017). Sulfate, ammonium, nitrate, dust, OA, and BC mass concentrations have been evaluated by surface observational networks of aerosol mass spectrometers (Zhang et al., 2007), the Interagency Monitoring of Protected Visual Environment (http://vista.cira.colostate.edu/improve) in the United States, and the European Monitoring and Evaluation Programme (http://www.emeap.int) in Europe. For most of these comparisons, simulations reproduced observed average concentrations within a factor of 2 (Matsui & Mahowald, 2017). BC mass concentrations and aerosol number concentrations (>10 nm in diameter) have been evaluated by various aircraft campaigns. The Aerosol Robotic Network (http://aeronet.gsfc.nasa.gov) and the Moderate Resolution Imaging Spectroradiometer (http://modis-atmos.gsfc.nasa.gov) have been used to evaluate aerosol optical depth and single scattering albedo. The ATRAS model has been shown to perform reasonably well in reproducing observed BC and non-BC mass and number concentrations as well as BC mixing states such as the fractions of BC-containing and BC-free particles and the coating amounts of BC-containing particles, including their size dependences (Matsui et al., 2013; Matsui, Koike, Kondo, Fast, et al., 2014). SOA concentrations observed in urban and suburban areas and over the outflow regions in East Asia have also been reproduced well by the ATRAS model (Matsui, Koike, Kondo, Takami, et al., 2014).
2.4. Definitions of Variables

BC DRE is defined based on the method of Ghan (2013) (for all-sky conditions) as the difference in the instantaneous shortwave radiative flux at the top of the atmosphere between cases considering all aerosol species and all aerosol species except BC. BC DRE is calculated for preindustrial, present-day, and future conditions, and the resulting BC DRF is calculated as the difference in BC DRE between present-day and preindustrial conditions. BC DRE and BC DRF are calculated for both all sources (fossil fuel + biofuel + biomass burning) and anthropogenic sources only (fossil fuel + biofuel) by tracking anthropogenic and biomass burning BC separately in the model.

BC DRE can be decomposed into several elements as shown in the following equations, where overbars denote global 5-year mean values:

\[
\text{DRE} = \overline{B} \times \text{NDRE}_B \\
= \overline{B} \times \overline{AAOD} \times \frac{\text{DRE}}{\overline{AAOD}} \\
= \overline{B} \times \text{MAC} \times \text{NDRE}_{AAOD} \\
= (E \times L) \times \left( \frac{\text{MAC}_{\text{core}}}{\overline{MAC}} \times \overline{E_{\text{abs}}} \right) \times \text{NDRE}_{AAOD}
\]

where \(B\) is the BC burden, \(E\) is BC emissions, \(L\) is BC lifetime, \(AAOD\) is absorption aerosol optical depth, \(MAC\) and \(MAC_{\text{core}}\) are mass absorption cross section of BC with and without coating species, respectively, \(E_{\text{abs}}\) is absorption enhancement defined by the ratio \(MAC/MAC_{\text{core}}\), and \(\text{NDRE}_B\) and \(\text{NDRE}_{AAOD}\) are normalized BC DRE with respect to \(B\) and \(AAOD\), respectively. Note that \(AAOD\) and related variables (\(MAC\), \(MAC_{\text{core}}\), and \(E_{\text{abs}}\)) are calculated at the wavelength of 550 nm, whereas DRE and DRF are integrated over all shortwave wavelengths. All variables in Equation 1 except \(MAC_{\text{core}}\) and \(E_{\text{abs}}\) are calculated from online simulations of the CAM5-chem/ATRAS model. \(MAC_{\text{core}}\) is calculated off-line by excluding non-BC species to isolate mass (or volume) and number concentrations of BC in individual bins (Matsui, Hamilton, & Mahowald, 2018), and \(E_{\text{abs}}\) is calculated from offline calculations of \(MAC\) and \(MAC_{\text{core}}\). Online and offline calculations of \(MAC\) agree well, as shown by Matsui, Hamilton, and Mahowald (2018).

### 3. Results

#### 3.1. BC DRF

The global burden of all aerosol species simulated by the CAM5-chem/ATRAS model is generally within the ranges of previous modeling studies (supporting information Table S1). The global mean BC DRE (from all sources) is estimated to be 0.071 and 0.36 W m\(^{-2}\) under preindustrial and present-day conditions, respectively, using CMIP6 emissions (Table 1), which yields global average BC DRF value of 0.29 W m\(^{-2}\) (Figure 1a). BC DRF is as large as approximately 2 W m\(^{-2}\) over BC source regions (e.g., East Asia and central Africa) and their surroundings (Figure 1a). When only anthropogenic (fossil fuel and biofuel) sources are considered, BC DRE is 0.0089 and 0.24 W m\(^{-2}\) under preindustrial and present-day conditions, respectively, and BC DRF is 0.23 W m\(^{-2}\) (Figure 1b). This BC DRF value is consistent with the multimodel mean value of 0.23 W m\(^{-2}\) (1750–2010) in the AeroCom Phase II experiments (Myhre, Samset, et al., 2013). The anthropogenic BC DRF in this study is also similar to that in a previous study using CMIP6 emissions (0.23 W m\(^{-2}\) for
the period 1750–2014 in Lund et al. (2018) when a MAC of 10 m$^2$ g$^{-1}$ is assumed). Recent observationally constrained estimates of BC DRF are both higher and lower than the unconstrained (model only) estimates (Chung et al., 2012; Q. Wang, Jacob, et al., 2014, R. Wang et al., 2016; Samset et al., 2014; X. Wang, Heald, et al., 2014).

Simulations using CMIP5 emissions produce lower values of BC DRE and BC DRF than those using CMIP6 emissions because present-day anthropogenic emissions (in 2010 for CMIP6 and 2000 for CMIP5) in CMIP6 are about 20% greater and preindustrial anthropogenic emissions (in 1750 for CMIP6 and 1850 for CMIP5) are about 50% lower than in CMIP5 (supporting information Table S2). For the CMIP5 case, BC DRF is estimated to be 0.18 W m$^{-2}$ for all sources and 0.13 W m$^{-2}$ for anthropogenic sources alone (supporting information Figure S3).

Compared with the multiple-mixing-state simulations, the single-mixing-state simulations yield higher BC DRF values (0.37 and 0.31 W m$^{-2}$ for all sources and anthropogenic sources, respectively, using CMIP6 emissions) because this representation overestimates absorption enhancement by assuming that all particles are internally mixed BC (Matsui, Hamilton, & Mahowald, 2018).

3.2. BC Absorption Efficiency

This section compares the BC variables in Equation 1 under preindustrial and present-day conditions. BC absorption efficiency (mainly MAC and $E_{abs}$) shows a large enhancement from preindustrial to present-day conditions. For all sources, the BC burden increases by a factor of 4.2 and BC DRE by a factor of 5.0 (Table 1), and $NDRE_B$ (the ratio of DRE to burden) is enhanced by 19%. Because the change in $NDRE_B$ is much clearer for anthropogenic BC (an enhancement of 73%), the next subsection focuses on anthropogenic BC.

3.2.1. $MAC_{core}$, $NDRE_{AAOD}$, and $E_{abs}$

In Equation 1, $NDRE_B$ can be decomposed into $MAC_{core}$, $NDRE_{AAOD}$, and $E_{abs}$. These three variables are enhanced from preindustrial to present-day conditions, as described below for anthropogenic sources.

First, $MAC_{core}$ is 9.8% higher under present-day than preindustrial conditions (5.8 and 5.2 m$^2$ g$^{-1}$, respectively; Table 1). This variable depends on the size distribution of BC cores (Matsui, Hamilton, & Mahowald, 2018). The mass median diameter of present-day BC cores (~190 nm) is slightly smaller than preindustrial BC cores (~220 nm), likely due to the larger present-day contribution of BC particles from fossil fuel combustion, which have a smaller median diameter at emission. Because the smaller median diameter of present-day BC produces a higher $MAC_{core}$ at a wavelength of 550 nm (see Figure 5a of Matsui, Hamilton and Mahowald (2018)), $MAC_{core}$ is slightly higher in the present-day simulations. Note that a reduction of $MAC_{core}$ at larger BC core diameters (>200 nm; Figure 5a of Matsui, Hamilton and Mahowald (2018)) is obtained because spherical BC particles are assumed in the optical property calculations. A recent study has suggested that $MAC_{core}$ is insensitive to particle size for large BC particles when a more complex fractal

Figure 1. Global distribution of black carbon (BC) direct radiative forcing (DRF) for (a) all sources (anthropogenic + biomass burning) and (b) anthropogenic sources alone. CMIP6 emissions for Years 2010 and 1750 were used to estimate BC DRF. Global-averaged BC DRF (W m$^{-2}$) between 1750 and 2010 are shown at lower left.
structure of BC particles is considered and that MACcore of large particles would be underestimated when spherical particles (Mie theory) are assumed (Forestieri et al., 2018).

Second, NDREAAOD is 12% higher under present-day than preindustrial conditions (148 and 132 W m\(^{-2}\), respectively). This variable is determined by the vertical distribution of BC (i.e., whether BC is located above clouds or not) and surface albedo (e.g., Samset & Myhre, 2015). Present-day NDREAAOD is 3–10% higher than preindustrial values at low latitudes (30°S to 30°N) and ~20% higher at high latitudes (>60°N) (not shown). The lengthening of BC lifetime in the simulations, from 4.3 days (preindustrial) to 4.6 days (present-day), may lead to slightly larger fractions of high-altitude BC and consequently higher NDREAAOD values in the present-day simulations. The BC lifetime is longer under present-day than preindustrial conditions (despite faster BC aging) because the diameter of BC-containing particles in the present day is smaller due to the greater contribution from fossil fuel sources (as noted above) and these smaller particles are activated less efficiently in clouds.

Third, \(E_{abs}\) is 41% higher under present-day than preindustrial conditions (1.5 and 1.1, respectively in Table 1). This variable is basically controlled by the amount of coating on BC-containing particles. The fraction of thickly coated large BC-containing particles (>200 nm in diameter with BC mass fractions between 0 and 0.4), which have high absorption enhancement (Matsui, 2016a), is larger for present-day than preindustrial conditions (Figure 2a and supporting information Figure S4). This enhancement of thickly coated BC fractions with time is much clearer for anthropogenic BC than total BC. Our previous study showed that thickly coated BC particles form mostly by coagulation of BC-containing and BC-free particles, whereas less coated BC particles form mainly by condensation (Matsui et al., 2013). The difference between preindustrial and present-day conditions in aging speed by coagulation is therefore likely an important cause of the historical enhancement of thickly coated BC fractions. In fact, aerosol number concentrations (>40 nm in dry diameter; \(N_{40}\)) have more than doubled over most of the Northern Hemisphere and increased by more than 5 times over source regions and their surroundings between 1750 and 2010 (Figure 2b). Observations have shown that most BC-containing particles from anthropogenic sources are fully coated, such that BC is not exposed at their surfaces (Moteki et al., 2014). This may be because both condensation and coagulation processes are usually fast over source and near-source regions, where concentrations of aerosols and their precursors are high. The shell-core treatment used in this study is therefore likely to be realistic.

Although thickly coated larger BC particles (>200 nm) form by coagulation (Myhre, Samset, et al., 2013), the hydrophobic-to-hydrophilic conversion of BC particles by aging processes is dominated by condensation because less coated (thinnly and moderately coated) smaller particles are produced more easily by condensation (Matsui, 2016b). The timescale of the hydrophobic-to-hydrophilic conversion and the contributions of
condensation and coagulation to aging processes (e.g., condensation is dominant during the day and coagulation is dominant during the night) estimated by Matsui (2016b) are generally consistent with other studies of BC aging processes (e.g., Fierce et al., 2015, 2017; Riemer et al., 2010).

3.2.2. MAC

Preindustrial and present-day MAC values, the product of $MAC_{\text{core}}$ and $E_{\text{abs}}$ in Equation 1, are estimated to be 7.7 and 9.0 m$^2$ g$^{-1}$, respectively (all sources). The global mean ratio of present-day to preindustrial MAC values (all sources) is 1.2, with the highest values around 1.4–1.6 over current source regions such as East Asia, Europe, North America, and North Africa (Figure 3a). The same ratio for anthropogenic BC is higher at 1.5 (Figure 3b) (preindustrial and present-day global mean values of 5.6 and 8.6 m$^2$ g$^{-1}$, respectively) because the ratio of present-day to preindustrial $E_{\text{abs}}$ values is higher for anthropogenic sources than for all sources (Figure 2a and Table 1).

As shown by Matsui, Hamilton and Mahowald (2018), MAC in the present-day simulations (9.0 m$^2$ g$^{-1}$ for all sources) is consistent with observed estimates of around 10 m$^2$ g$^{-1}$ (Cui et al., 2016; Ram & Sarin, 2009; Yttri et al., 2014; Zanatta et al., 2016) and the multimodel mean value of $\sim$8 m$^2$ g$^{-1}$ in the AeroCom models (Myhre, Samset, et al., 2013). The value of $E_{\text{abs}}$ in the present-day simulations (1.6) is also similar to previous estimates ($\sim$1.5; Fierce et al., 2016; Wu et al., 2018). The normalized BC DRE with respect to BC aerosol optical depth at 550 nm (not to be confused with $NDRE_{\text{AOD}}$ in Equation 1, which involves AAOD) is estimated to be 142 W m$^{-2}$ (for anthropogenic sources) in the present-day simulations in this study, a value similar to the multimodel mean value of 133 W m$^{-2}$ (range 84–216 W m$^{-2}$) in the AeroCom models (Myhre, Samset, et al., 2013).

Figure 3. Global distribution of the ratio of present-day (Year 2010) to preindustrial (Year 1750) BC mass absorption cross section (BC MAC) under the multiple mixing state representation for (a) all sources and (b) anthropogenic sources and under the single mixing state representation for (c) all sources and (d) anthropogenic sources. Global-averaged values are shown at lower left.
The enhancement in MAC from preindustrial to present-day conditions by aging processes is considerably smaller in the single-mixing-state simulations (Figures 3c and 3d) than in the multiple-mixing-state simulations (Figures 3a and 3b) for both all sources (1.9% vs. 17%) and anthropogenic sources (15% vs. 54%). Because the single-mixing-state representation cannot represent the increase in thickly coated BC particles by aging processes, MAC values are less sensitive to changes in emissions and aging processes between pre-industrial and present-day conditions than the multiple-mixing-state representation. These results demonstrate that emissions, number concentrations, and BC mixing state should be represented accurately when calculating the speed of BC aging processes and estimating BC absorption efficiency. Models that assume a constant MAC (or \( E_{abs} \)) or use a simple representation of BC aging processes may overestimate BC absorption efficiency in preindustrial times.

### 3.2.3. Impact on DRF Estimation

Here I evaluate how the historical enhancement in BC MAC affects the estimation of BC DRF. Defining \( DRE_s \) as present-day BC DRE for BC category \( s \) (all or anthropogenic sources) and \( DRE_s^* \) as the present-day BC DRE calculated with the preindustrial MAC value, the difference between \( DRE_s \) and \( DRE_s^* \) can be considered as the difference in BC DRF (\( \Delta DRF_s \)) due to the enhancement of MAC from preindustrial (PI) to present-day (PD) conditions as follows:

\[
\Delta DRF_s = DRE_s (PD) - DRE_s^* (PD) = DRE_s (PD) - DRE_s (PD) \times \frac{MAC_s (PI)}{MAC_s (PD)}
\]

The \( \Delta DRF \) value is calculated for both total and anthropogenic BC. The global mean \( \Delta DRF \) for all sources is 0.051 W m\(^{-2}\) (Figure 4a), corresponding to 18% of BC DRF (0.29 W m\(^{-2}\)), and the global mean \( \Delta DRF \) for anthropogenic sources is 0.086 W m\(^{-2}\) (Figure 4b), or 37% of anthropogenic BC DRF (0.23 W m\(^{-2}\)). The change in aging speed from preindustrial times to the present day therefore can increase BC DRF by 20–60%. The increase is greater than 0.1 W m\(^{-2}\) near-source regions and greater than 0.5 W m\(^{-2}\) in East Asia (eastern China, Figure 4), showing that faster aging of BC today than in the past can enhance BC DRF substantially, both regionally and globally.

### 3.3. Importance of Non-BC Emissions and Secondary Aerosol Formation

Simulations for the Year 2100 based on the RCP4.5 emissions scenario yield a MAC value of 8.7 m\(^2\) g\(^{-1}\) (all sources), which is close to the present-day MAC value (8.8 m\(^2\) g\(^{-1}\)) and higher than the preindustrial value (7.5 m\(^2\) g\(^{-1}\)). Because \( N_{so} \) and secondary aerosol burden differ considerably between these simulations, non-BC emissions and secondary aerosol formation may largely account for the different MAC values in these simulations. The importance of non-BC species to BC absorption enhancement has been evaluated by both observational/experimental studies (e.g., D. Liu et al., 2017) and modeling studies (e.g., Jacobson, 2001; Stier et al., 2006).
To explore this topic, two sensitivity simulations were performed, one in which RCP4.5 emissions of all aerosols and their precursor species other than BC, dust, and sea salt were doubled and one in which they were halved. Although BC emission flux does not change, MAC varies within a range of 7.7–9.7 m² g⁻¹ in the two sensitivity simulations (red line at 4 Tg year⁻¹ in Figure 5a). In the doubled simulation, N₄₀ and secondary aerosol concentrations (sulfate + nitrate + SOA) at the surface are enhanced from 352 to 450 cm⁻³ and from 1.3 to 3.3 μg m⁻³, respectively, whereas these concentrations fall to 270 cm⁻³ and 0.50 μg m⁻³ in the halved simulation. Values of MAC increase monotonically with respect to secondary aerosol burden (Figure 5b), consistent with a recent observational/theoretical study (D. Liu et al., 2017). This result shows that the differences of MAC between the preindustrial, present-day, and future simulations can be explained by differences in non-BC emissions and secondary aerosol formation rates. BC DRE for RCP4.5 2100 has a range of 0.13–0.18 W m⁻² in these simulations (Figure 5c). In a comparison with future emission scenarios of the Shared Socioeconomic Pathways (SSPs) (O’Neill et al., 2014), global emissions of OA, sulfur dioxide (SO₂), and nitrogen oxides (NOₓ) in the halved simulation are generally similar to those in SSP1-1.9 (Year 2100) (Lund et al., 2019). Emissions in the doubled simulation are similar to those in SSP3-7.0 for OA and 30–40% lower than SSP3-7.0 for SO₂ and NOₓ. The spread of secondary aerosol burden, BC MAC, and BC DRE in 2100 in the SSP scenarios therefore will be larger than those in the doubled and halved simulations of this study. This suggests that BC MAC and BC DRE in the future climate are sensitive to future changes in non-BC (OA, SO₂, and NOₓ) emissions and to the choice of future emission scenarios.

Doubled and halved sensitivity simulations were also performed for the present-day condition (2010). The range of MAC values in these simulations (8.1–9.8 m² g⁻¹, red line at 10 Tg year⁻¹ in Figure 5a) is similar to the range in the RCP4.5 2000 sensitivity simulations (Figures 5a and 5b). BC DRE is 0.31 and 0.42 W m⁻² in the halved and doubled simulations, respectively (Figure 5c), although BC emission flux is unchanged. Compared with the base simulation, BC MAC and NDREₐₓₐₒₐₒ are enhanced by 9.4%, and 5.9%, respectively, in the doubled simulation, which lead to a 18% increase in BC DRE (results are similar in simulations with the RCP4.5 2000 emissions). BC DRE in the halved simulation is 13% lower than the base simulation due to the reduction of MAC and NDREₐₓₐₒₐₒ by 9.4% and 4.0%, respectively. These results demonstrate that amounts and changes of secondary aerosols and their precursor emissions play a key role in estimating BC absorption efficiency, BC DRE, and BC DRF.

The enhancement of secondary aerosols in the doubled simulation increases anthropogenic BC AAOD by 10–20% at low and middle latitudes and decreases it by 5–10% at high latitudes (Figure 6b). This change can be explained as the result of greater absorption enhancement by faster secondary aerosol formation, which is dominant at low and middle latitudes, and more efficient wet removal, which reduces BC transport to polar regions (Figure 6a). This result is qualitatively consistent with the finding of Stier et al. (2006) that enhanced hygroscopicity counteracts the effect of enhanced BC absorption. In the simulations shown here, the enhancement of absorption is more important than the shortened BC lifetime in terms of global BC AAOD changes (Figure 6).
3.4. Sensitivity to Emission Variables

Five additional simulations were conducted to investigate the sensitivity of BC absorption efficiency and BC DRE to emission variables assumed in this study (such as size distributions and mixing states in emissions and emission fluxes in the preindustrial conditions). The simulations are listed in Table 2, and their results are summarized in Figure 7.

In the base simulations described in sections 3.1–3.3, the number median diameters of emissions were assumed to be 70 nm for fossil fuel sources and 100 nm for biofuel and biomass burning sources (section 2.2). However, current global aerosol models differ considerably in their treatment of emission size distributions (Mann et al., 2014). Based on the uncertainty range of emission size distributions estimated in previous studies (Carslaw et al., 2013; Lee et al., 2013), two simulations were made with the upper and lower limit of the uncertainty range: The number median diameters of fossil fuel and biofuel/biomass burning emissions are 30 and 50 nm, respectively, for the Small-size case and 80 and 200 nm, respectively, for the Large-size case (Table 2). The mass fluxes of aerosol emissions in the Small-size and Large-size cases are the same as the base case, whereas the number flux in the Small-size case is greater than that in the Large-size case. Therefore, the Small-size case can be expected to yield faster BC aging processes, longer BC lifetime, and larger BC DRE (Matsui, Hamilton, & Mahowald, 2018). Total (anthropogenic) BC DRF (1750–2010) is 0.41 W m$^{-2}$ (0.35 W m$^{-2}$) for the Small-size case and 0.21 W m$^{-2}$ (0.18 W m$^{-2}$) for the Large-size case (Figure 7). In the Small-size case, total (anthropogenic) BC MAC and $E_{abs}$ are 8.5% (33%) and 11% (36%) higher under present-day than preindustrial conditions. In the Large-size case, total (anthropogenic) BC MAC and $E_{abs}$ are 70% (164%) and 21% (74%) higher under present-day conditions. The preindustrial to present-day enhancement of BC MAC and $E_{abs}$ is smaller in the Small-size case because aging processes in preindustrial simulations are fast enough to produce high BC MAC and $E_{abs}$ values (9.7 m$^2$ g$^{-1}$ and 1.6, respectively, for total BC). These results show that BC MAC and $E_{abs}$ and their preindustrial to present-day changes are highly sensitive to the treatment of emission size distributions.

Table 2

| Simulation     | Emission number median diameter (nm) | Emission mixing state (pure BC percentage) | Emission years and flux |
|----------------|--------------------------------------|-------------------------------------------|-------------------------|
| Large size     | 80, 200                               | 100%                                      | 1750 and 2010           |
| Small size     | 30, 50                                | 100%                                      | 1750 and 2010           |
| Ext_half       | 70, 100                               | 50%                                       | 1750 and 2010           |
| BVOC × 1.2     | 70, 100                               | 100%                                      | 1750 (BVOC × 1.2)       |
| BB × 2         | 70, 100                               | 100%                                      | 1750 (BB × 2)           |

Note. Number median diameters are shown for fossil fuel emissions (left) and biofuel and biomass burning emissions (right). The geometric standard deviation of emission size distributions is 1.8 for all sources and all simulations. In the Ext_half simulation, half of BC emissions are assumed to be pure BC (externally mixed BC) and the other half are assumed to be mixed BC and OA (internally mixed BC). Shell-to-core diameter ratios of internally mixed BC emissions are assumed to be 1.1 for fossil fuel sources and 1.4 for biofuel and biomass burning sources. BVOC, biogenic volatile organic compounds; BB, biomass burning.
One sensitivity simulation (Ext_half) examined the treatment of mixing states in emissions. Similar to simulations by Matsui, Hamilton, and Mahowald (2018), the simulation assumed that half of BC is emitted as pure BC and the other half as internally mixed BC (BC-OA mixture). Because the mixing state distribution of BC emissions is not well known, the total diameter to BC diameter (shell-to-core) ratio of internally mixed BC particles was assumed to be 1.1 for fossil fuel emissions and 1.4 for biofuel and biomass burning emissions based on limited observations (Kondo et al., 2010, 2011). Because half of BC was already mixed with OA in emissions, MAC and E_{abs} values are 20–60% higher than those in the base simulation, which assumed that all BC is emitted as pure BC (Figure 7). MAC and E_{abs} are 24% and 20% higher, respectively, under present-day than preindustrial conditions for anthropogenic BC. These are lower than the values in the base simulations (54% for MAC and 41% for E_{abs}) because internally mixed BC emissions lead to higher MAC and E_{abs} values (8.9 m² g⁻¹ and 1.5, respectively, for anthropogenic BC) in the preindustrial simulations. These results show that a better understanding of BC mixing state distributions near emission sources and their changes from preindustrial to present-day climate will be important for improved estimation of BC absorption efficiency. The shell-to-core diameter ratio assumed here for internally mixed BC emissions currently has a large uncertainty. Reducing this uncertainty and determining a realistic mixing state distribution of BC emissions will be necessary for accurate BC simulations.

Aerosol emissions from natural sources in preindustrial times are also highly uncertain. The last two sensitivity simulations focused on biogenic and biomass burning emissions. SOA formation from biogenic emissions in preindustrial times can be a source of uncertainty in BC aging processes and absorption enhancement. Because some studies have estimated that global emissions of biogenic volatile organic compounds (BVOC) are about 10–20% higher in preindustrial time than the present day (e.g., Acosta Navarro et al., 2014; Hantson et al., 2017), a simulation was done with 20% enhanced preindustrial isoprene and monoterpane emissions (BVOC × 1.2). This simulation yields a 7.0% higher OA burden than the base simulation, and the impact of this change on MAC and E_{abs} (for both total and anthropogenic BC) is very small (Figure 7). Although this simulation did not consider the preindustrial to present-day changes in the spatial distribution of biogenic emissions, it suggests that BC absorption efficiency is not sensitive to preindustrial biogenic emissions because of the relatively small change in BVOC emissions from preindustrial time to present day.

In the base simulation, preindustrial biomass burning emissions are about 15% lower than present-day emissions, but a recent study suggests that biomass burning emissions in preindustrial times may be much larger than current estimates (Hamilton et al., 2018). A sensitivity simulation with doubled preindustrial biomass burning emissions (BB × 2) enhances preindustrial MAC and E_{abs} for total BC by 10% (from 7.7 to 8.5 m² g⁻¹) and 7% (from 1.4 to 1.5), respectively, whereas changes in MAC and E_{abs} for anthropogenic BC are almost negligible (Figure 7). This result shows that the absorption efficiency of anthropogenic BC is not sensitive to preindustrial biomass burning emissions.

In addition to the factors considered above, other climate factors not considered in this study may influence the differences in BC simulations comparing preindustrial, present-day, and future climates. For example, changes of ocean and sea ice surface conditions may influence atmospheric circulation and hydrological...
cycles and may change the BC burden and distribution. Changes of temperature and long-lived climate forcers may affect chemical reaction rates and oxidation capacity in the atmosphere and the resulting aerosol formation and BC aging processes. Evaluations of their importance to BC absorption efficiency and radiative forcing may be important in future studies.

4. Summary

This study evaluated BC absorption efficiency (e.g., MAC and absorption enhancement) under preindustrial and present-day conditions by using a size- and mixing-state-resolved global aerosol model. BC DRF from preindustrial times to present day was 0.29 and 0.23 W m$^{-2}$ for all sources and anthropogenic (fossil fuel + biofuel) sources, respectively, in simulations using CMIP6 emissions (and 0.18 and 0.13 W m$^{-2}$ for all sources and anthropogenic sources, respectively, in simulations using CMIP5 emissions).

The present-day normalized BC DRE with respect to BC burden ($NDRE_{DP}$) was found to be enhanced relative to preindustrial values by 19% and 73% for all sources and anthropogenic sources, respectively. MAC, the major driver of the differences between preindustrial times and present day, was enhanced by 17% for all sources and 54% for anthropogenic sources because the historical increase in aerosol aging speed increases the fraction of thickly coated BC particles, which have higher absorption enhancement. The impact of the historical enhancement of MAC on BC DRF was estimated to be 0.051–0.086 W m$^{-2}$ globally (reaching a maximum of >0.5 W m$^{-2}$ near-source regions in East Asia), suggesting that the historical change in aging speed during industrial times has increased BC DRF by 20–60%. It should be emphasized that these effects can be simulated only in models that consider BC mixing state with sufficient resolution. These results show that a better understanding of BC aging processes in various environments, based on both observations and model simulations, will be important for accurate estimation of BC DRF.

BC DRF and MAC are sensitive to non-BC emissions and secondary aerosol formation. In sensitivity simulations with doubled and halved emissions for all aerosols and their precursor species (other than BC, dust, and sea salt), present-day MAC and BC DRE values varied within the ranges of 8.1–9.8 m$^2$ g$^{-1}$ and 0.31–0.42 W m$^{-2}$, respectively, although BC emission flux was unchanged. This can explain why the BC MAC varies in different emission data sets. Sensitivity simulations also showed that BC DRF and MAC are sensitive to aerosol size distributions and mixing states in emissions, whereas they are not sensitive to natural emission fluxes in preindustrial time because of the relatively small change in their emissions (10–20%) from preindustrial time to present day. This study demonstrated that BC absorption efficiency is highly sensitive not only to BC emissions but also non-BC emissions, secondary aerosol formation, and aerosol microphysical properties in emissions through their influence on aging processes and the resulting formation rate of thickly coated BC particles. This conclusion suggests that improving the understanding and representation of particle size, mixing state, non-BC emission flux, and secondary aerosol formation in addition to BC emission flux will be necessary to make more accurate estimates of BC absorption efficiency, DRE, and DRF in preindustrial, present-day, and future climates.

Conflict of Interest

The author declares no conflicts of interest.

Data Availability Statement

The CESM/CAM model can be obtained from the website (http://www.cesm.ucar.edu/). Data used in this study are available from the following link (http://doi.org/10.5281/zenodo.3991105). For further questions regarding the CAM-ATRAS model and its data, contact the corresponding author.

References

Abdul-Razzak, H., & Ghan, S. J. (2000). A parameterization of aerosol activation: 2. Multiple aerosol types. Journal of Geophysical Research, 105(D5), 6837–6844. https://doi.org/10.1029/1999JD901161

Abdul-Razzak, H., & Ghan, S. J. (2002). A parameterization of aerosol activation: 3. Sectional representation. Journal of Geophysical Research, 107(D3), 4026. https://doi.org/10.1029/2001JD000443
Jacobson, M. Z. (2001). Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols. Nature, 409(6821), 695-697. https://doi.org/10.1038/35055518

Jacobson, M. Z., Turco, R. P., Jensen, E. J., & Toon, O. B. (1994). Modeling coagulation among particles of different composition and size. Atmospheric Environment, 28(7), 1327–1338. https://doi.org/10.1016/1352-2310(94)90928-1

Koch, D., Balkanski, Y., Bauer, S. E., Easter, R. C., Furtak, S., Ghan, S. J., et al. (2011). Soot microphysical effects on liquid clouds, a multi-model investigation. Atmospheric Chemistry and Physics, 11(3), 1051–1064. https://doi.org/10.5194/acp-11-1051-2011

Matsui, H., Koike, M., Takegawa, N., Kajino, M., et al. (2011). Emissions of black carbon, organic, and inorganic aerosols from biomass burning in North America and Asia in 2008. Journal of Geophysical Research, 116, D08204. https://doi.org/10.1029/2010JD015152

Matsui, H., Koike, M., Takegawa, N., Kajino, M., et al. (2011). Comparison and traceability of black carbon measurements made by laser-induced incandescence, thermal-optical transmittance, and filter-based photo-absorption techniques. Aerosol Science and Technology, 45(2), 295–312. https://doi.org/10.1080/02786826.2010.532115

Kulmala, M., Lehtinen, K. E. J., & Laaksonen, A. (2006). Cluster activation theory as an explanation of the linear dependence between formation rate of 3 nm particles and sulphuric acid concentration. Atmospheric Chemistry and Physics, 6(3), 787–793. https://doi.org/10.5194/acp-6-787-2006

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., et al. (2010). Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application. Atmospheric Chemistry and Physics, 10(15), 7017–7039. https://doi.org/10.5194/acp-10-7017-2010

Kondo, Y., Matsui, H., Moteki, N., Sahu, L., Takegawa, N., Kajino, M., et al. (2011). Anthropogenic aerosol microphysical properties among AeroCom models of a range of complexity. Atmospheric Chemistry and Physics, 11(12), 4909–4931. https://doi.org/10.5194/acp-11-4909-2011

Kondo, Y., Matsui, H., Takegawa, N., Kajino, M., et al. (2011). Anthropogenic aerosol microphysical properties among AeroCom models of a range of complexity. Atmospheric Chemistry and Physics, 11(12), 4909–4931. https://doi.org/10.5194/acp-11-4909-2011

Kondo, Y., Matsui, H., Takegawa, N., Kajino, M., et al. (2011). Anthropogenic aerosol microphysical properties among AeroCom models of a range of complexity. Atmospheric Chemistry and Physics, 11(12), 4909–4931. https://doi.org/10.5194/acp-11-4909-2011

Lund, M. T., Myhre, G., Haslerud, A. S., Skeie, R. B., Griesfeller, J., Platt, S. M., et al. (2018). Concentrations and radiative forcing of anthropogenic aerosols from biomass burning in North America and Asia in 2008. Journal of Geophysical Research: Atmospheres, 123, 1808–1821. https://doi.org/10.1002/2015JD023999

Matsui, H. (2016a). Black carbon simulations using a size- and mixing-state resolved three-dimensional model: 1. Radiative effects and their uncertainties. Journal of Geophysical Research: Atmospheres, 121, 1793–1807. https://doi.org/10.1002/2015JD023998

Matsui, H. (2016b). Black carbon simulations using a size- and mixing-state resolved three-dimensional model: 2. Aging timescale and its impact over East Asia. Journal of Geophysical Research: Atmospheres, 122, 1808–1821. https://doi.org/10.1002/2015JD023999

Matsui, H. (2017). Development of a global aerosol model using a two-dimensional sectional method: 1. Model design. Journal of Advances in Modeling Earth Systems, 9, 1921–1947. https://doi.org/10.1002/2017MS001090

Matsui, H., Hamilton, D. S., & Mahowald, N. M. (2018). Black carbon radiative effects highly sensitive to emitted particle size when resolving mixing-state diversity. Nature Communications, 9(1), 3446. https://doi.org/10.1038/s41467-018-05635-1

Matsui, H., Koike, M., Kondo, Y., Fast, J. D., & Takigawa, M. (2014). Development of an aerosol microphysical module: Aerosol two-dimensional bin module for formation and Aging Simulation (ATRAS). Atmospheric Chemistry and Physics, 14(18), 10,315–10,331. https://doi.org/10.5194/acp-14-10315-2014

Matsui, H., Koike, M., Kondo, Y., Moteki, N., Fast, J. D., & Zveri, R. A. (2013). Development and validation of a black carbon mixing state resolved three-dimensional model: Aging processes and radiative impact. Journal of Geophysical Research: Atmospheres, 118, 2304–2326. https://doi.org/10.1002/2012JD018446

Matsui, H., Koike, M., Kondo, Y., Takami, A., Fast, J. D., Kanaya, Y., & Takigawa, M. (2014). Volatility basis-set approach simulation of organic aerosol formation in East Asia: Implications for anthropogenic-biogenic interaction and controllable amounts. Atmospheric Chemistry and Physics, 14(18), 9513–9535. https://doi.org/10.5194/acp-14-9513-2014

Matsui, H., Koike, M., Takegawa, N., Wiedensohler, A., Fast, J. D., & Zveri, R. A. (2011). Impact of new particle formation on the concentrations of aerosols and cloud condensation nuclei around Beijing. Journal of Geophysical Research, 116, D19208. https://doi.org/10.1029/2011JD016025

Matsui, H., & Mahowald, N. (2017). Development of a global aerosol model using a two-dimensional sectional method: 2. Evaluation and sensitivity simulations. Journal of Advances in Modeling Earth Systems, 9, 1887–1920. https://doi.org/10.1002/2017MS001097

Matsui, H., Mahowald, N. M., Moteki, N., Hamilton, D. S., Ohtani, S., Yoshida, A., et al. (2018). Anthropogenic combustion iron as a complex climate forcer. Nature Communications, 9(1), 1593. https://doi.org/10.1038/s41467-018-03997-0

Morrison, H., & Gettelman, A. (2008). A new two-moment bulk stratiform cloud microphysics scheme in the Community Atmosphere Model, version 3 (CAM4). Part I: Description and numerical tests. Journal of Climate, 21(5), 3642–3659. https://doi.org/10.1175/2008JCLI2105.1

Moss, R. H., Edmonds, J. A., Hibbard, K. A., Manning, M. R., Rose, S. K., van Vuuren, D. P., et al. (2010). The next generation of scenarios for climate change research and assessment. Nature, 46/7282), 747–756. https://doi.org/10.1038/nature08823
Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H., Ulbrich, I., et al. (2007). Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes. Geophysical Research Letters, 34, L13801. https://doi.org/10.1029/2007GL029979

References From the Supporting Information

Adams, P. J., Seinfeld, J. H., & Koch, D. M. (1999). Global concentrations of tropospheric sulfate, nitrate, and ammonium aerosol simulated in a general circulation model. Journal of Geophysical Research, 104(D11), 13,791–13,823. https://doi.org/10.1029/1999JD900083

Bauer, S. E., Koch, D., Unger, N., Metzger, S. M., Shindell, D. T., & Streets, D. G. (2007). Nitrate aerosols today and in 2013: A global simulation including aerosols and tropospheric ozone. Atmospheric Chemistry and Physics, 7(19), 5043–5059. https://doi.org/10.5194/acp-7-5043-2007

Bellouin, N., Rae, J., Jones, A., Johnson, C., Haywood, J., & Boucher, O. (2011). Aerosol forcing in the Climate Model Intercomparison Project (CMIP5) simulations by HadGEM2-ES and the role of ammonium nitrate. Journal of Geophysical Research, 116, D20206. https://doi.org/10.1029/2011JD016074

Feng, Y., & Penner, J. E. (2007). Global modeling of nitrate and ammonium: Interaction of aerosols and tropospheric chemistry. Journal of Geophysical Research, 112, D01304. https://doi.org/10.1029/2006JD007135

Huijard, P., Balkanski, Y., & Schulz, M. (2014). A global model simulation of present and future nitrate aerosols and their direct radiative forcing of climate. Atmospheric Chemistry and Physics, 14(20), 11,031–11,063. https://doi.org/10.5194/acp-14-11031-2014

Huneeus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Prospero, J., Kinne, S., et al. (2011). Global dust model intercomparison in AeroCom phase I. Atmospheric Chemistry and Physics, 11(15), 7781–7816. https://doi.org/10.5194/acp-11-7781-2011

Liao, H., Seinfeld, J. H., Adams, P. J., & Mickley, L. J. (2004). Global radiative forcing of coupled tropospheric ozone and aerosols in a unified general circulation model. Journal of Geophysical Research, 109, D16207. https://doi.org/10.1029/2003JD004456

Pringle, K. J., Tost, H., Messasse, S., Stell, B., Giannadaki, D., Nenes, A., et al. (2010). Description and evaluation of GMXe: A new aerosol submodel for global simulations (v1). Geoscientific Model Development, 3(2), 391–412. https://doi.org/10.5194/gmd-3-391-2010

Rodriguez, M. A., & Dabdub, D. (2004). IMAGES-SCAPE2: A modeling study of size- and chemically resolved aerosol thermodynamics in a global chemical transport model. Journal of Geophysical Research, 109, D02203. https://doi.org/10.1029/2003JD004369

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., et al. (2006). Analysis and quantification of the diversities of aerosol life cycles within AeroCom. Atmospheric Chemistry and Physics, 6(7), 1777–1813. https://doi.org/10.5194/acp-6-1777-2006

Tsagaridis, K., Daskalakis, N., Kanakidou, M., Adams, P. J., Artaxo, P., Bahadur, R., et al. (2014). The AeroCom evaluation and intercomparison of organic aerosol in global models. Atmospheric Chemistry and Physics, 14(19), 10,845–10,895. https://doi.org/10.5194/acp-14-10845-2014

Xu, L., & Penner, J. E. (2012). Global simulations of nitrate and ammonium aerosols and their radiative effects. Atmospheric Chemistry and Physics, 12(20), 9479–9504. https://doi.org/10.5194/acp-12-9479-2012