Improved Simulations of Global Black Carbon Distributions by Modifying Wet Scavenging Processes in Convective and Mixed-Phase Clouds

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Abstract
In-cloud wet scavenging dominates the wet removal of aerosols in the atmosphere, but is not well represented in climate models. Aircraft measurements of black carbon (BC) concentrations suggest that models commonly overestimate BC concentrations in the upper troposphere of the tropics by more than one order of magnitude but underestimate BC burdens in polar latitudes. In this study, we improved the in-cloud wet scavenging parameterizations for convective clouds and mixed-phase clouds to better characterize BC abundances in the remote atmosphere (remote oceans and polar regions) with a global model, CAM5-ATRAS2. The modified wet scavenging processes in the model achieved a more realistic simulation of BC concentrations over both the tropics and the Arctic. The new, unified scheme for vertical transport and wet removal during deep convection generally reproduced the observed low mixing ratios (about 0.1 ng kg⁻¹) of BC in the middle and upper troposphere over the tropics, and the Wegener–Bergeron–Findeisen process (WBF) lowered the wet removal efficiency of BC from mixed-phase clouds and consequently increased BC burdens in the Arctic by about a factor of 2. The BC direct radiative forcings increased by 20% globally (from 0.26 to 0.31 W m⁻²), and more importantly by a factor of 2 in the Arctic (from 0.09 to 0.18 W m⁻²). Our results indicated that good agreement between modeled and observed BC concentrations could be obtained in the remote atmosphere without requiring the relatively short global BC lifetime (~4 days) suggested by previous studies.

1. Introduction
Atmospheric black carbon (BC), which originates primarily from the combustion of biomass and fossil fuels, plays an important role in climate change by altering the global radiation budget through its strong absorption of solar radiation, its interactions with clouds, and reductions of the albedo of snowpack and ice in the Arctic (Flanner et al., 2007; Jacobson, 2001; Koch & Del Genio, 2010). Atmospheric three-dimensional models have been widely used to quantify the impacts of BC on the global climate because observational data are insufficient to capture the temporal and spatial variations of BC concentrations on a global scale (e.g., Schulz et al., 2006). However, large uncertainties exist in model simulations of BC abundances, especially in the remote atmosphere. Compared to available observations, many models tend to underestimate BC concentrations in the Arctic, especially during the winter and early spring (Koch et al., 2009; J. Liu et al., 2011), and hence they cannot reproduce the observed seasonal cycles of surface BC concentrations. In the middle and upper troposphere of the tropical Pacific, the modeled BC concentrations are generally overestimated by more than one order of magnitude when compared with the observations from the HiPER Pole-to-Pole Observations (HIPPO) aircraft campaigns (Schwarz et al., 2010, 2013).

Wet scavenging processes have been proposed to explain the model bias of BC simulation in the remote atmosphere (Kipling et al., 2013, 2016; J. Liu et al., 2011; Q. Wang et al., 2014), because they significantly determine the lifetime and global distribution of BC, but are not well represented in current, advanced global climate models (GCMs). Specifically, BC wet removal is dominated by in-cloud wet scavenging, which refers to the activation of BC-containing particles into cloud droplets and further scavenging by precipitation. After being emitted into the atmosphere, BC can be gradually coated with highly hygroscopic components like sulphate and nitrate aerosols (aging processes), which can then serve as cloud condensation nuclei (CCN) (Bond et al., 2013; Moteki et al., 2007). In GCMs, the in-cloud wet removal rate of BC particles is usually calculated as a function of the activated fraction of BC, the cloud fraction in a grid, and the cloud water to rainwater conversion rate. Simple adjustments of in-cloud scavenging efficiency in models by tuning some...
related parameters have not been found to yield better simulations of BC mass across all latitudes (Bond et al., 2013). For example, an enhancement of wet scavenging rates (that is, a reduction of BC lifetimes) may reduce the over-prediction of BC in the middle and upper troposphere over tropical regions, but would exacerbate the low bias of BC in the Arctic (Garrett et al., 2010). Comprehensive treatments that improve in-cloud wet scavenging parameterizations over different regions are thus needed to reduce the model bias of BC on a global scale.

For tropical and sub-tropical regions, in-cloud wet scavenging associated with deep convection is a significant determinant of the aerosol abundances and their vertical profiles in the region (Ervens, 2015). In a standard GCM, like the Community Atmosphere Model, the separate treatment of the vertical transport and wet removal of aerosols in convective clouds may lead to too much transport of aerosols aloft during convective updrafts (X. Liu et al., 2012). H. Wang et al. (2013) and Yu et al. (2019) proposed a unified scheme that treats these two processes simultaneously and considers the secondary aerosol activation from entrained air above the cloud base, which can efficiently remove BC during deep convection in the middle and upper troposphere in the tropics. How this unified scheme behaves on a global scale, however, remains unclear. It appears that the inclusion of secondary aerosol activation during deep convective transport in the model would exacerbate the low bias of the simulated BC concentrations in the temperate and polar latitudes (Yu et al., 2019).

The fact that models commonly underestimate BC loadings in the air by more than a factor of 2 in the Arctic during the haze season (winter and early spring) has impeded our understanding of BC-radiation interactions and BC effects on snow albedo in the region (Eckhardt et al., 2015; Koch et al., 2009). As reported by Qi, Li, Li, et al. (2017) and J. Liu et al. (2011), the causes of the underestimates of Arctic BC in models include the uncertainties in BC emissions in Russia, treatments of BC aging, and the representation of dry and wet deposition. Wet scavenging processes have been suggested to be the dominant factor responsible for the seasonality of BC concentrations in the Arctic because they impact both the transport efficiency of BC from midlatitude source regions and the local wet removal (Mahmood et al., 2016; Shen et al., 2017). Our recent studies have demonstrated that the treatments of maximum supersaturation in liquid clouds significantly influence the aging rates and CCN activities of BC-containing particles and could alter the surface BC concentrations in the Arctic by up to one order of magnitude in winter and early spring (Matsui & Moteki, 2020; Moteki et al., 2019). Browse et al. (2012) have shown that the seasonal transition from high BC concentrations in winter to much lower concentrations in summer in the Arctic is controlled by the seasonality of scavenging by ice-phase clouds and warm cloud. A strong suppression of BC scavenging in ice clouds would cause the modeled seasonal cycle of BC concentrations to be consistent with observations. It is well documented that mixed-phase clouds are predominantly distributed in the mid- and high-latitudes, and they can affect the wet scavenging of BC via the so-called Wegener–Bergeron–Findeisen process (WBF) (Fan et al., 2012). The WBF refers to the transfer of water vapor from liquid to ice phases when the vapor pressure is between the saturation vapor pressure over ice and water droplets (Bergeron, 1935). This process leads to evaporation of the aerosol materials in the droplets back into the interstitial air and subsequently reduces the rates of aerosol scavenging in mixed-phase clouds (Cozic et al., 2007; Verheggen et al., 2007). Qi, Li, He, et al. (2017) have demonstrated that the simulation of BC in air and snow in the Arctic is substantially improved by including the effects of the WBF on BC wet removal.

In this study, we aim to improve the simulation of BC vertical profiles in both tropics and polar regions by modifying the aerosol in-cloud wet scavenging parameterizations in convective clouds and mixed-phase clouds. Recent studies have examined the effects of one or the other of those processes on BC concentrations to address the model bias at targeted latitudes (Fan et al., 2012; Qi, Li, Li, et al., 2017; Yu et al., 2019), but the combined effects have never been reported, and it remains unclear how these processes impact BC burdens and radiative effects on a global scale. Our results provide important implications for modeling global BC distributions and lifetimes.

2. Methods

2.1. Model Setups

We use the Community Atmospheric Model version 5 (CAM5) (X. Liu et al., 2012) with the Aerosol Two-dimensional bin module for foRmation and Aging Simulation version 2 (CAM5-chem/ATRAS2)
The ATRAS2 module uses a two-dimensional sectional representation with 12 particle size bins (from 1 to 10,000 nm in diameter) and 8 BC mixing state bins (from fresh BC to aged BC-containing particles) to simulate various microphysical and chemical processes of aerosols, including new particle formation, condensation/coagulation, aerosol activation, wet/dry deposition, and interactions with radiation and clouds. The chemical aging of BC-containing particles is explicitly considered in the module using the method of Matsui et al. (2013).

The model was run at a horizontal resolution of 1.9° × 2.5° with 30 vertical layers from the surface to ∼40 km on a global scale. Eight parallel simulations (4 × 2) were performed. The simulations included both preindustrial and present-day conditions, and for each of those conditions, we considered four different treatments of in-cloud wet scavenging processes (described in Section 2.2) to demonstrate their effects on BC distributions and the radiative forcings. The global emissions of aerosols and their precursors in 1750 (preindustrial) and 2010 (present-day) were taken from Hoesly et al. (2018) based on the Community Emissions Data System for anthropogenic emissions and from van Marle et al. (2017) for biomass burning emissions. All simulations in this study were run with present-day climatological data (i.e., the ocean and ice surface boundary and long-lived climate forcers including CO₂, CH₄, and other greenhouse gases). We performed simulations for the years 2008–2011, with the last 3 years used for analysis. The meteorological fields for each year were nudged by using the Modern-Era Retrospective analysis for Research and Applications Version 2 data. Note that the preindustrial simulations were used only for probing the impacts of model treatments on BC radiative forcings in this study. We calculated the BC all-sky shortwave radiative effect due to aerosol-radiation interactions following the methods in Ghan (2013). The BC direct effect due to aerosol-radiation interaction was calculated online as the difference between the standard radiation fluxes and the diagnosed fluxes, for which BC was subtracted in the radiation module.

To evaluate the performances of the modified model, we used comprehensive measurements of BC concentrations on a global scale, including aircraft campaigns from HIPPO (Wofsy, 2011), Atmospheric Tomography Mission (ATom) (Wofsy, 2018), NASA-DC8 over North America (Toon et al., 2016), and A-FORCE over East Asia (Oshima et al., 2012), and two in situ surface stations in the Arctic (Sinha et al., 2017). The HIPPO data are obtained from five deployments across the central Pacific during 2009–2011 and include measurements of BC mass concentrations in the troposphere with a single-particle soot photometer (SP2) instrument (Schwarz et al., 2013). The ATom data contain four deployments covering the Pacific and Atlantic during 2016–2018. There were totally nine deployments covering Pacific regions and the Arctic. Observed BC concentrations at each deployment of HIPPO and ATom were averaged across 12–14 altitude bins at about 1-km intervals. For comparison with aircraft measurements, we averaged the monthly mean simulated vertical BC profiles from the surface to the upper troposphere over four latitude bins across the central Pacific and three bins across the Atlantic Ocean (i.e., box averaging). In addition, we used the monthly mean BC concentrations observed at two Arctic surface sites, Barrow (71.3°N, 156.6°W) and Ny-Ålesund (78.9°N, 11.9°E) (Sinha et al., 2017).

### 2.2. Improvements of In-Cloud Wet Scavenging

We considered the improvements of in-cloud wet scavenging in convective clouds and mixed-phase clouds and made four parallel simulations (namely BASE, CONV, WBF, and CONV_WBF as listed in Table 1) to examine their effects on BC distributions and radiative forcings. Other physical and chemical processes including below-cloud wet scavenging and precipitation evaporation were the same among the four cases following the standard configuration of CAM5-ATRAS2 (Matsui, 2017). For the BASE case in our model, vertical transport and wet removal during deep convection were treated separately (i.e., in different modules), without involving the aerosol activation from entrained air during convective updraft. The convective in-cloud scavenging rate (SRCC) was calculated as follows:

\[
SRCC = CLDC\cdot FRACP\cdot F\cdot ACT\cdot TRACER
\]
In which CLDC denotes the fraction of convective clouds in one grid, FRACP denotes the transfer rate of cloud water to rainwater, F_ACT represents the constant fraction of interstitial aerosols activated into cloud droplets, and TRACER denotes concentrations of interstitial aerosols in the air. The constant F_ACT was set to either 0, 0.4, or 0.8 for BC-containing particles with different diameters and mixing states that corresponded to aerosol critical supersaturation values of >1.0, 0.5–1.0, and ≤0.5%, respectively (Moteki et al., 2019). Environmental supersaturation was not treated for sub-grid convective clouds in our model. The other parameters in Equation 1 are online calculated in the model. However, the vertical transport of aerosols during deep convection was treated in another module that did not involve wet removal.

In the CONV simulation, we replaced the original treatment of in-cloud convective wet scavenging with a unified scheme that combined the deep convective transport and wet removal in the same module, so that aerosols within the convective updraft can be efficiently removed from the atmosphere with precipitation, which can then reduce the quantity of aerosols aloft. The mass-continuity equations for the unified deep convection scheme have been proposed by H. Wang et al. (2013) and Yu et al. (2019) and are shown in Equations 2 and 3 below:

\[
\frac{\partial (M_u q_{ud})}{\partial p} = E_u q_e - D_u q_{ad} - \text{Act} \cdot q_{ad} \tag{2}
\]

\[
\frac{\partial (M_u q_{uc})}{\partial p} = E_u q_{ec} - D_u q_{uc} + \text{Act} \cdot q_{ad} - \text{Wet} \cdot q_{uc} \tag{3}
\]

where \(M_u\) denotes the air mass velocity in convective updraft; \(E_u\) and \(D_u\) denote rates of entrainment and detrainment, respectively, within the updraft; \(q_{ad}\) and \(q_{uc}\) denote the interstitial and cloud-borne aerosol concentrations, respectively, within the updraft; \(q_e\) and \(q_{ec}\) denote the grid-mean interstitial and cloud-borne aerosol concentrations, respectively; and \(\text{Act}\) and \(\text{Wet}\) denote the activation rate (1/s) for interstitial aerosols that have been entrained into convective updraft and the conversion rate of convective cloud water to rainwater, respectively. \(\text{Act}\) is set to 0.04 in accord with Yu et al. (2019) without the consideration of BC aging processes. Note that in the BASE case, the activation fractions (F_ACT values) were defined for grid-cell aerosols and differed from the \(\text{Act}\). Following H. Wang et al. (2013), we assumed that the secondary aerosol activation happened only in the updraft and that the cloud-borne aerosols from the detrained air could be resuspended as interstitial aerosols.

The physical processes of WBF in stratiform, mixed-phase clouds have been treated in the BASE model, but their effects on aerosol removal have not been considered, which could overestimate the removal efficiency of aerosols. A parameterization of WBF effects on aerosol removals is proposed by Cozic et al. (2007) based on the aerosol and cloud measurements at Jungfraujoch, Switzerland, a continental background site. The BC scavenging rate \(r_{scav}\) in stratiform mixed-phase clouds is expressed by Equation 4:

\[
r_{scav} = \min \left(0.06 + 1.07 \cdot \exp\left(-9.33 \sqrt{\text{IMF}}\right), 1.0\right) \tag{4}
\]

where IMF is the ice mass fractions (0–1) in clouds. In this study, the WBF effects were limited to the temperature ranges 248–261K and 265–273K, as suggested by (Qi, Li, He, et al., 2017), to exclude the influence of the riming process in mixed-phase clouds.

3. Results and Discussion

3.1. Improvements of BC Simulations

Figure 1 compares modeled and observed monthly BC concentrations within four latitude bins and five HIPPO campaigns covering different seasons. In the tropical region (20°S–20°N), the BASE simulation (blue line in Figure 1) severely overestimated the BC mass in the middle and upper troposphere by up to a factor of 10. The modeled BC concentrations generally fell within the range 1–10 ng kg\(^{-1}\) at pressures < 700 hPa, whereas most of the observed concentrations were within 0.1–1 ng kg\(^{-1}\) during all five deployments. Here in the BASE simulation, we assumed a constant fraction of BC particles was activated into cloud droplets.
Figure 1. Comparison of simulated vertical profiles of BC mass concentrations from four model experiments with observations (mean + standard deviations) from five HIPPO deployments in different latitude bands across the central Pacific (160°E–140°W). BC, black carbon; HIPPO, HIAPER Pole-to-Pole Observations.
from the surface layer to the upper troposphere via convective wet scavenging processes; and the vertical transport by deep convection is treated separately. Even though some fractions of BC were removed from the atmosphere by convective precipitation, there was still a considerable amount of BC lifted into the upper troposphere in the model. Similarly, the ensemble of AeroCom phase II (including the standard version of CAM5.1) produced an order-of-magnitude overestimation of BC from the model ensemble in the remote troposphere over the tropics (Schwarz et al., 2013).

To reduce the model bias, we applied a unified convective wet-scavenging scheme (CONV) that took into consideration secondary aerosol activation above the cloud base during deep convection. The CONV simulation (orange lines in Figure 1) satisfactorily captured the low levels of the mixing ratios (0.1 ng kg⁻¹) of BC in the upper troposphere of the tropical region; most of the modeled BC concentrations fell within the observed ranges of the mean values ± standard deviations. In this case, BC-containing particles were efficiently removed by convective precipitation during updraft transport and could not be lifted into the upper atmosphere, as pointed out by Yu et al. (2019) and H. Wang et al. (2013). Over the tropics, the vertically integrated BC burdens at pressures < 700 hPa dropped by an average of 68% during five HIPPO periods in the CONV simulation, and the resulting BC vertical profiles generally agreed better with the observations than in the BASE simulation. The Pearson correlation coefficient between the simulated and observed vertical profiles averaged over five deployments increased from 0.22 (BASE) to 0.51 (CONV). These results demonstrated that the new deep convection scavenging treatment in the model yielded a more realistic BC profiles. The CONV simulation also improved the magnitude or shape of simulated BC profiles in the middle and upper troposphere over the 20°–60°N region by lowering the convective transport of BC aloft. However, in the South Hemisphere mid-latitude region (20°–60°S), the new deep convection scheme caused the BC concentrations to be underestimated in the upper troposphere.

To address the underestimation of BC in the Arctic, we included the WBF effects on the in-cloud wet scavenging of aerosols within mixed-phase clouds in the model (the WBF case), and we used the HIPPO data to examine how this process impacted BC simulations in different latitude bands. First, in the Arctic region (60°–90°N), the WBF simulation (green lines in Figure 1) significantly reduced the model underestimation in spring (HIPPO3) and winter (HIPPO1 and HIPPO2); the absolute mean bias for vertically integrated BC concentrations shrank from −6.1 to −12 ng kg⁻¹ in the BASE case to −0.95 to −4.3 ng kg⁻¹ in the WBF case in those periods. Such large increases in BC loadings in the Arctic were caused by the large reduction in BC scavenging efficiency in mixed-phase clouds because of WBF effects, and more BC was therefore released from liquid clouds into the interstitial air. During the summer periods (HIPPO4 and HIPPO5), both the BASE and WBF cases overestimated BC concentrations. At midlatitudes in the Northern Hemisphere (20°N–60°N), the modeled BC concentrations with WBF effects were a little biased high in the middle troposphere (300–700 hPa) during the HIPPO 1 and 2 periods, while during HIPPO3 the absolute mean bias of the vertically integrated BC was reduced from −20 to −4.7 ng kg⁻¹. The WBF effect was less important in the tropics and resulted in only a slight increase (∼1 ng kg⁻¹) of BC concentrations during HIPPO measurement periods.

Overall, comparison of the CONV_WBF simulation with the HIPPO data revealed that the improved convective wet scavenging scheme dominated the changes of BC concentration in the tropics (except for HIPPO4) and improved the shape and magnitude of the BC profiles in the middle and upper troposphere, whereas the WBF effect substantially enhanced simulated BC loadings in the Arctic, which agreed better with the observed BC profiles during the winter and spring (HIPPO1-3). Temperate latitude regions were subject to the joint effects of CONV and WBF processes, which partially counteracted one another, and the resulting performances of BC simulations varied among different the HIPPO deployments. In addition, a comparison of simulated BC vertical profiles with other aircraft measurements—including ATom in both the Pacific and Atlantic regions, A-FORCE over East Asia, and NASA-DC8 over North America (note that the targeted years differed between our simulations and these datasets)—further demonstrated that the overall performance achieved with the CONV and WBF improvements was good (Figures S1 and S2).

A comparison with the surface BC measurements at two stations in the Arctic shows that our simulation in the BASE case severely underestimated those concentrations in spring and winter (Figure 2). The inclusion of WBF effects increased the surface BC mass by about a factor of 2 and effectively reduced the model bias, but the gaps were still significant. In addition to WBF effects, many other factors can determine the
seasonal cycle of simulated BC burdens in the Arctic. First, the aging processes of BC particles and associated activation fractions into cloud droplets control their transport efficiency (J. Liu et al., 2011; Matsui & Moteki, 2020; Shen et al., 2017). Slower aging processes have been found to enhance BC burdens in the Arctic during winter and spring and substantially reduce the underestimation there. Second, the representation of BC wet removal efficiency in liquid and mixed-phase clouds is another key factor driving the BC seasonal cycle. Browse et al. (2012) have suggested that in-cloud wet scavenging of BC would need to be much more efficient in warm clouds than in ice clouds to reproduce the observed seasonal distribution. Moreover, neglect or underestimation of gas-flaring emissions around the Arctic would also lead to the underestimation of BC concentrations (Qi, Li, Li, et al., 2017; Stohl et al., 2013).

Furthermore, in line with previous studies (Samset et al., 2014), we averaged the data from all five HIPPO deployments to represent an approximate annual mean level. As shown in Figure 3, the ratios of the corresponding annual mean simulated BC concentrations from the BASE, CONV, WBF, and CONV_WBF cases to the HIPPO observations were calculated separately at different latitudinal and vertical bins. Notably, ratios greater than 10 in the upper troposphere at latitudes of 20°S–20°N for the BASE case demonstrated a large overestimation of simulated BC. Those ratios dropped to 0.5–2.5 with the revised deep convection scheme (CONV). The WBF process resolved the large underestimation of BC concentrations in the middle and low troposphere of the Arctic region (60°–90°N), where the ratios increased from 0.1–0.5 to 0.5–1.0. The CONV_WBF scheme represented the combined effects of these two different processes, and the ratios of BC mass were closer to 1.0 than the BASE ratios at most latitudes. The CONV_WBF case showed an overall better performance on a global scale than that considering the CONV or WBF process alone. We therefore suggest that the improved in-cloud wet scavenging schemes can effectively reduce the overall range of discrepancies relative to the ensemble of HIPPO data, especially for the tropics and Arctic regions.

Most of our model results over different latitude bins were close to the mean or median values of the multi-model ranges of profiles from the AeroCom simulations (Schwarz et al., 2013, 2010) and within the corresponding 25th and 75th percentile ranges. With the exception of the tropics, the simulated BC profiles in the middle and upper troposphere in the CONV_WBF/CONV cases were beyond the lower bound of the AeroCom results and were in much better agreement with HIPPO measurements. It should be mentioned that inclusion of these two new processes in GCMs cannot ideally improve the BC profiles over all latitude bands and seasons. Other factors like emissions, model resolution, and model data sampling methods (flight-tracked or box-averaging methods) may also influence the comparison between model results and aircraft observations to some degree. Moreover, the model improvements themselves were also subject to some limitations. Because the current model lacks enough information to diagnose the environmental supersaturation in convective clouds, we used the same activation rates for BC particles from the entrained air above the cloud base, despite their diversity of mixing state and size bins. This simple treatment may introduce uncertainties in the wet removal efficiency of BC mass and associated vertical transport flux. More detailed model treatments of both environmental supersaturation and activation rates linked to chemical aging of BC particles in sub-grid convective clouds are warranted in future studies. The parameterization of WBF effects on BC wet scavenging was based on an in-situ measurement at one mountain site (Cozic et al., 2007).
which may be limited for modeling BC on a global scale. Despite these potential limitations, the new in-cloud wet scavenging processes are suggested to be physically more realistic than the default treatment.

3.2. Changes in Global BC Distribution and Lifetime

In this section, we investigate the differences of the modeled BC concentrations between the BASE and the physically improved simulations (CONV, WBF, and CONV_WBF) to elucidate on the quantitative effects of the modified in-cloud scavenging parametrizations on the global spatial distribution and lifetime of BC in the model. Figure 4 illustrates the pressure-latitude zonal mean distribution of BC mass concentrations during 2009–2011 in the BASE simulation and the ratios of the improved model results to the BASE case. The hotspots of BC zonal mean concentrations (>0.1 µg m⁻³) were captured by the model at northern temperate latitudes (20°–60°N) and around the tropics (Figure 4a), which are caused primarily by intensive fossil fuel emissions and biomass burning, respectively. Our simulations demonstrated that the CONV improvement

Figure 3. Ratios (R) of the simulated to observed BC concentrations during HIPPO deployments at different altitudes and latitude bins. The averages of five HIPPO deployments were used for the calculation of the ratios. Note that R < 1 denotes underestimation by our simulation and R > 1 denotes overestimation. BC, black carbon; HIPPO, HIAPER Pole-to-Pole Observations.
decreased the BC burdens in the middle and upper troposphere by factors of 1.5–10, respectively, from the tropics to the polar regions, and moderately elevated the BC loading in the low troposphere at temperate and tropical latitude bins by 10%–50%. We interpret these results in the following way.

Figure 4. Latitude-pressure distribution of annual mean BC concentrations for the BASE simulation (a) and the BC mass ratios of CONV_WBF to BASE (b), CONV to BASE (c), and WBF to BASE (d). BC, black carbon.
First, the improved in-cloud wet scavenging parameterization for deep convection led to efficient removal of aerosol within the convective updraft when precipitation occurred, which impeded the upward transport of BC and thereby reduced their concentrations in the upper layers. This process is different from that of Allen and Landuyt (2014), who have indicated that excessive BC aloft in the tropics could be attributable to overestimation of convective mass fluxes in models. Moreover, the reductions of BC in the middle and upper troposphere also weakened the transport efficiency of BC to the polar regions and decreased the burden there. Second, there was an increase in BC concentrations in the low troposphere (>700 hPa) around the tropics in the CONV simulation, which agreed with the results of H. Wang et al. (2013) but differed from those of Yu et al. (2019). These differences in previous studies may be attributable to the treatment of BC wet scavenging during deep convection in the original models as well as the treatment of secondary aerosol activation in the new scheme. In the BASE simulation, constant activation fractions and the fractional area of convective clouds were used to represent the transformation of interstitial aerosols to cloud borne aerosols to calculate the scavenging amounts of BC. This was distinct from the new, unified scheme applied in the CONV case in which the activated fractions were highly dependent on BC in the entrained air during updraft. Our results imply that the constant fractions used for BC activation in convective clouds in the BASE model may be too high to scavenge BC in the lower troposphere of the tropics. We also carried out a sensitivity simulation for deep convective removal in the BASE case by halving those fixed activation fractions (\( F_{ACT} \) in Equation 1) of BC. It shows that the BC concentrations in the low troposphere were almost same as in the CONV case, whereas the positive bias of the simulated BC in the upper levels was exacerbated (Figure S3).

Unlike the CONV simulation, the WBF effects released BC from cloud droplets to interstitial air when the mixed-phase clouds were present and the vapor pressure lies between the saturation vapor pressure of the liquid and ice phases. This process reduced the in-cloud scavenging efficiency of BC. Substantial increases (i.e., WBF/BASE > 1.5) in ambient BC concentrations were predominately found at polar latitudes from surface to the upper troposphere and within the mid-upper troposphere at temperate latitudes (Figure 4d), where the low temperature and relatively high frequency of mixed-phase clouds results in strong WBF effects compared to other regions. In the Arctic, the annual mean BC concentrations increased by a factor of 1.5 near the surface to more than a factor of 3 in the middle troposphere (~400 hPa) due to the reductions of BC in-cloud wet scavenging in mixed-phase clouds, which were similar to the results in Qi, Li, He, et al. (2017). The percentage increases of BC concentrations in the temperate latitudes fell mostly within the range 50%–250% at pressures < 700 hPa, and this favored the poleward transport of BC and further enhanced the loading in the polar regions. The joint effects of WBF and CONV partially counteracted one another in the mid-upper troposphere over the middle and polar latitudes (Figure 4b). It’s clear to see that the net changes in the tropics were dominated by the CONV effects, whereas those in the polar latitudes were dominated by the WBF. In addition, the impact of modified convective wet scavenging on BC concentrations had little longitudinal dependence, while the WBF effects appeared to be more appreciable in the remote Pacific region (Figure S4). The BC particles over the remote Pacific Ocean experienced much more wet removal by precipitation during long-range transport than those in source regions and could be more susceptible to changes of wet removal processes (Matsui & Moteki, 2020).

The impacts of CONV and WBF on BC burdens exhibited distinct seasonal characteristics (Figure 5). In the BASE case, the distributions of the zonal mean BC burdens were bimodal for the DJF, JJA, and SON periods (Figure S5). The BC burdens peaked at temperate latitudes (around 30°N) throughout the year because of the tremendous anthropogenic emissions from East and South Asia. In the tropical regions, high BC burdens were associated with intensive biomass burning events in middle and southern Africa. The CONV and WBF effects had seasonal distinctions in altering the BC loading over different latitudes bands. First, the improved in-cloud wet scavenging scheme for deep convection redistributed the BC vertical gradients and associated burdens, especially in the middle and upper troposphere (corresponding to altitude above 3 km in the study). The BC loadings above 3 km were reduced by about 40% throughout the year in the tropics without significant seasonal variation, because deep convection was frequent in all seasons. At the temperate latitudes of North Hemisphere (20°–60°N), the reductions were as large as 40%–44% during JJA and SON periods, but were much smaller during DJF and MAM (22%–26%), because deep convection in this region is mainly concentrated in the summer with strong air instability. The similar seasonality of the CONV effect was apparent at the latitudes 20°–60°S in the South Hemisphere; the reductions in BC burdens were...
Figure 5. Changes in zonal mean BC burdens in the CONV, WBF, and CONV_WBF simulations relative to the BASE. The BC burdens were integrated for (a–d) all vertical levels and (e–h) above 3 km at five latitude bands (90°–60°S, 60°–20°S, 20°S–20°N, 20°–60°N, and 60°–90°N) and during four seasons, that is, DJF (December, January, and February), MAM (March to May), JJA (June to August), and SON (September to November), respectively. BC, black carbon.
primarily found above 3 km during DJF and MAM seasons (summer). In contrast, the WBF effects were dominant under colder air conditions (e.g., polar latitudes, upper troposphere, and winter season). The BC burdens in the Arctic (60°–90°N) substantially increased due to the WBF process (Figures 5a–5d), showing the greatest changes in winter and spring (+120%) and the smallest in summer (+40%). For temperate and tropical latitudes, the WBF effect was only pronounced at high altitudes during the winter and spring. The joint effects of the CONV and WBF differed among latitudes and seasons. Importantly, the WBF effect dominated the changes in BC concentrations in the Arctic and during winter time at temperate latitudes, while the CONV effects were more apparent in the tropics.

We now discuss the consistency and differences between our improvements and previous studies with similar updated cloud microphysical processes. As shown in this study and previous ones, models tend to severely overestimate the BC loading in the upper troposphere of the tropics, and implement of more efficient scavenging in convective clouds is therefore suggested (Kipling et al., 2013; Q. Wang et al., 2014; Zhang et al., 2020). For instance, although Q. Wang et al. (2014) have included nucleation scavenging of BC for ice nuclei at $T < 258 \text{ K}$ and impaction scavenging of hydrophobic BC aerosols in convective updraft, the model overestimation of BC concentrations over the tropics was still significant. Allen and Landuyt (2014) varied the in-cloud convective wet removal efficiency in the standard CAM5 model by increasing the solubility factor of BC from 0.4 to 0.9 and found that the global BC loading above 500 hPa decreased by 34%. In contrast, this BC loading in our case (CONV) decreased by 78%. The convective scheme analogous to our study by Kipling et al. (2013), which treated the vertical transport and wet removal simultaneously, has shown much improved BC vertical distributions in climate models. Our results, combined with previous studies, thus suggest that a tight coupling between transport and wet scavenging in convective clouds can efficiently reduce overestimation of BC aloft in models. In addition, the representation of in-cloud scavenging in mixed-phase and ice clouds are of importance, particularly for high latitudes and the upper troposphere. Xu et al. (2019) have reported that including the BC heterogeneous ice nucleation via immersion freezing reduces BC concentrations in the middle and upper troposphere (<600 hPa) across all latitudes by up to one order of magnitude. This reduction tends to narrow the model bias in the tropics but may worsen the underestimation of BC in the Arctic. The role of BC in ice nucleation has been poorly understood until now (Kanji et al., 2020). Croft et al. (2010) have indicated that, in addition to nucleation, in-cloud impaction scavenging of BC is significant in cold clouds at high latitudes. In the current study, we did not consider the impacts of ice nucleation and impaction scavenging processes on the abundance of BC particles, but we added the effect of WBF on BC wet scavenging in mixed-phase clouds. That effect increased the BC budget in the Arctic by about a factor of 2 and simulated the BC vertical profiles reasonably well.

Table 2 summarizes the differences in the global inter annual mean BC burdens and lifetimes among different cases and the results of previous studies. Compared to the BASE case, the improved deep convection scheme increased the BC burdens slightly from 0.24 to 0.25 mg m$^{-2}$. As noted above, the new scheme sharply reduced BC concentrations in the upper troposphere, but at the same time increased BC loading in the lower troposphere. The WBF effect resulted in a large increase of BC burden (from 0.24 to 0.28 mg m$^{-2}$) due to its reductions in BC scavenging efficiency in mixed-phase clouds. The joint effect of the CONV and WBF was an increase of ~20% (from 0.24 to 0.29 mg m$^{-2}$) in the global mean BC burden. The estimates of BC burdens in these four cases fell within the range of previous studies (e.g., AeroCom Phase II and other single-model studies), among which there are large differences in both input emissions and model treatments of wet scavenging processes (Kipling et al., 2016).

Furthermore, we used the global mean BC lifetime, defined as the global mean BC burden divided by the annual emissions as a metric of the removal efficiency of modeled BC from the atmosphere to compare our results with other studies. Corresponding to the changes in BC burdens, the lifetime extends from 4.8 to 5.7 days (Table 2), which resulted in better agreement with the aircraft observations in the central Pacific. Previous studies (Bauer et al., 2013; Q. Wang et al., 2014) have argued that a shorter lifetime (around 4 days) due to stronger wet removal is required in models to reproduce the observed concentrations in the remote atmosphere from HIPPO campaigns. However, our results suggest that by improving the representation of the in-cloud wet scavenging in deep convection, the GCM shows good agreement with measured BC at high altitudes in tropical regions without shortening the global-mean BC lifetime. This improvement resulted from the inclusion of a more realistic wet scavenging scheme for convective clouds that treats the vertical
transport and wet removal simultaneously, instead of simply tuning the wet scavenging parameters in models. It’s also important to note that if our model obtains a shorter BC lifetime through faster BC aging and stronger wet removal, the model underestimation of BC in the lower and middle troposphere of the Arctic will be exacerbated. Lund et al. (2018) found that a global-mean lifetime of 5.5 days with a range of 5.3–5.9 days is associated with the lowest model bias for modeling BC in the Arctic based on comparisons with several datasets of aircraft observations, which is quite consistent with our studies (5.7 days). We therefore conclude that, to improve the model agreement with the aircraft measurements across different latitudes, especially in the mid- and upper troposphere of tropical regions, a shorter global BC lifetime (e.g., <4 days) is not necessary and the optimum global-mean lifetime may be 5–6 days.

### 3.3. Impacts on BC AAOD and Radiative Forcings

We further investigated the impacts of improved wet scavenging schemes on the BC absorption aerosol optical depth (AAOD) and instantaneous radiative forcings due to aerosol-radiation interactions (RFari). The changes in zonal mean BC AAOD were strongly dependent on the variations in BC burdens among the four model cases (Figure 6a). Compared to the BASE simulation, the BC AAOD in the CONV simulation

| References             | Emissions (Tg yr⁻¹) | Burden (mg m⁻²) | Burden (Tg) | Lifetime (day) | RFari (W m⁻²) |
|------------------------|---------------------|-----------------|-------------|----------------|---------------|
| BASE                   | 9.5                 | 0.24            | 0.12        | 4.8            | 0.26          |
| CONV                   | 9.5                 | 0.25            | 0.13        | 5.0            | 0.24          |
| WBF                    | 9.5                 | 0.28            | 0.14        | 5.5            | 0.33          |
| CONV_WBF               | 9.5                 | 0.29            | 0.15        | 5.7            | 0.31          |
| Schulz et al. (2006)   | 6.3                 | 0.25 ± 0.08     | –           | 7.3 ± 2.3      | 0.25 ± 0.08   |
| Jacobson (2012)        | 9.3                 | 0.18            | 0.089       | 3.2            | –             |
| Myhre et al. (2013)    | –                   | 0.08–0.31       | –           | 3.5–17.1       | 0.18 ± 0.07   |
| Bond et al. (2013)     | 17                  | 0.55            | –           | –              | 0.71          |
| H. Wang et al. (2013)  | 7.7                 | –               | 0.083–0.17  | 3.9–7.9        | –             |
| Q. Wang et al. (2014)  | 6.5                 | 0.15            | –           | 4.2            | 0.19          |
| Qi, Li, He, et al. (2017) | 8.5               | 0.35            | –           | 8.0            | –             |

Abbreviations: BC, black carbon; RFari, radiative forcings due to aerosol-radiation interactions.

Figure 6. Comparison of latitudinal mean distributions of BC AAOD (a) and RFari (b) averaged over 2009–2011 for the four BASE, CONV, WBF, and CONV_WBF simulations. BC, black carbon; AAOD, absorption aerosol optical depth; RFari, radiative forcings due to aerosol-radiation interactions.
increased by 13% in the tropics, which is dominated by the increased BC loadings in the lower troposphere, while the BC AAOD decreased by 33% in the Arctic because of decreases of BC concentrations in middle and upper troposphere. The WBF effects increase BC AAOD globally, with the most significant enhancement (by more than a factor of 2) in the Arctic. Overall, the global-mean BC AAOD for the CONV_WBF was 0.0026, or 21% higher than that in the BASE. Despite these changes, our results generally fell within the range of previous estimates (Jacobson, 2012; Schulz et al., 2006; Q. Wang et al., 2014). It should be noted that BC AAOD in the CONV_WBF run from CAM5-chem/ATRAS2 was still underestimated when compared to AERONET observations on the global scale as we discussed previously (Matsui & Mahowald, 2017). Bond et al. (2013) suggested doubling the global BC emissions to 17 Tg yr\(^{-1}\) in order to match the AERONET AAOD, and their modified mean AAOD was 0.006. In addition, the absorption enhancement of radiation by BC coatings associated with the mass absorption cross section of BC is another important factor that determines AAOD and remains a very uncertain parameter in climate models (Bond et al., 2013; Matsui, 2020; Matsui et al., 2018; Stier et al., 2006).

We calculated the annual mean BC RF\(_{\text{ari}}\) by comparing the simulations between preindustrial (1750) and present-day (2010) conditions (Figures 6b and Table 2). In line with the variations of AAOD, the joint effects of CONV and WBF increases the global mean BC RF\(_{\text{ari}}\) from 0.26 (BASE) to 0.31 W m\(^{-2}\) (+19%). The responses of BC RF\(_{\text{ari}}\) to the treatments of in-cloud wet scavenging were most pronounced in the Arctic, mainly because of the WBF effects. The increase of BC RF\(_{\text{ari}}\) by a factor of 2 from 0.09 to 0.18 W m\(^{-2}\) indicates greater warming effects of BC in the Arctic atmosphere. Our results suggest that BC burdens and radiative effects in the Arctic were very sensitive to aerosol activation and removal in large-scale clouds, which has also been demonstrated in our recent study (Matsui & Moteki, 2020). We note that the global mean RF\(_{\text{ari}}\) was lower in the CONV simulation than in the BASE (0.24 vs. 0.26 W m\(^{-2}\) ), though the BC AAOD was slightly higher in the former (0.0023 vs. 0.0022). Our improved convective wet scavenging scheme reshaped the BC vertical profile and reduced BC concentrations in the remote atmosphere (e.g., middle and upper troposphere) by up to one order of magnitude, and because the radiation forcing efficiency (defined as the ratio of RF to AAOD) of BC particles increases with altitude, the RF\(_{\text{ari}}\) was more sensitive to the variation of BC concentrations at higher altitudes (Samset & Myhre, 2015; Zarzycki & Bond, 2010). This could be true for the altitude dependence of BC direct radiative forcing, but the semi-direct effects induced by BC particles that were not considered in this study have been recently suggested to largely counteract the direct forcing at almost all altitude (Samset & Myhre, 2015).

Spatially, the annual mean BC RF\(_{\text{ari}}\) in the BASE simulation was as large as 2 W m\(^{-2}\) over BC source regions, such as East Asia, South Asia, and Central Africa, as well as their outflow regions (Figure 7a). In the CONV case, because the unified convective scheme reduced BC loadings in the middle and upper troposphere, there were widespread negative changes (up to \(-0.2\) W m\(^{-2}\) in source regions) of BC RF\(_{\text{ari}}\) that were distributed mainly in the North Hemisphere (Figure 7b). The occurrences of enhanced BC forcings in the tropics was attributable to the increased BC loadings in the lower atmosphere compared to the BASE (Figure 4). The WBF effects increase the BC RF\(_{\text{ari}}\) by more than +0.1 W m\(^{-2}\) in most of the North Hemisphere and by as much as +0.5 W m\(^{-2}\) in East Asia. Overall, the positive changes in BC RF\(_{\text{ari}}\) in the CONV_WBF case are dominated by the WBF processes, indicating greater BC warming effects via the direct interaction with radiations.

### 4. Conclusion

Recent studies have suggested that current global models greatly overestimate BC concentrations in the middle and upper troposphere over the tropics and underestimate BC loadings in the Arctic. These biases have been attributed to insufficient model representation of in-cloud wet scavenging. However, simply reducing or increasing BC scavenging rates cannot yield better performances at all latitudes. In this study, we improved the treatment of aerosol in-cloud scavenging processes in convective clouds and mixed-phase clouds in a global climate model (CAM5-chem/ATRAS2) based on the constrain from comprehensive aircraft measurement datasets on a global scale.

This study indicates that the modified wet scavenging processes can achieve a more realistic simulation of BC concentrations over both the tropics and the Arctic. Similar to previous studies, the standard simulation
The CONV simulation with a unified scheme for convective transport and wet removal reproduce the low mixing ratios (close to 0.1 ng/kg) of observed BC for the tropics, because the new scheme considered the secondary aerosol activation from the entrained air during updraft and efficiently removed BC-containing particles during convective transport from the surface to the upper troposphere. The inclusion of WBF effects increased the BC burden in the Arctic by a factor of two and consequently reduced the model underestimation of BC based on the HIPPO and ATom measurements. The ratios of simulated to observed BC concentrations for the lower and middle troposphere of the Arctic increased from 0.1 in the BASE case to above 0.5 in the CONV_WBF case. Overall, the CONV effects were dominant in the tropics, and the WBF effects were dominant in polar regions. At the temperate latitudes, the combined effects of these two processes partially counteracted one another within the mid-upper troposphere. It's also interesting to find that the impacts of the new convective scheme on BC burdens were particularly apparent during the summer time at temperate latitudes, while WBF effects were more important during the winter and spring due to the cold air conditions, especially in the Arctic. The global-mean BC AAOD was 0.0026 in the CONV_WBF case, 21% higher than in the BASE case. The inclusion of CONV and WBF increased the global mean BC RFari from 0.26 (in the BASE case) to 0.31 W m$^{-2}$. Specifically, the doubled BC loadings in the Arctic dominated by the WBF process increased the regional-mean RFari by a factor of 2 (from 0.09 to 0.18 W m$^{-2}$), which indicates greater warming effects of BC in the Arctic atmosphere.

Overall, the more realistic in-cloud wet scavenging schemes than the default treatments could effectively reduce the magnitude of the discrepancies between model results and the HIPPO measurements, especially in the tropics and Arctic regions. We conclude that, to improve the agreement between modeled BC and

**Figure 7.** Global distribution of annual mean BC RFari in the BASE simulation (a) and their changes for the CONV (b), WBF (c), and CONV_WBF (d) simulations. BC, black carbon; RFari, radiative forcings due to aerosol-radiation interactions.
the aircraft measurements along BC vertical profiles, especially in the mid- and upper troposphere of the tropics, a shorter BC lifetime (e.g., around 4 days) suggested by previous studies is not necessary and the optimum global-mean lifetime may be 5–6 days.

Data Availability Statement

The averaged vertical profiles of BC from five HIPPO deployments were provided by U.S. National Oceanic and Atmospheric Administration (NOAA) and the original data can be accessed via NCAR/EOL (https://www.eol.ucar.edu/field_projects/hippo). The BC aircraft measurements from ATom campaigns can be accessed from NASA Earthdata (https://daac.ornl.gov/ATOM/campaign). NASA-DC8 measurement data over North America are available at http://www.wair.larc.nasa.gov/missions/seac4rs/index.html for the SEA-C4RS mission and http://www-air.larc.nasa.gov/cgi-bin/ArchView/dc3 for the DC3 mission.

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