High Sensitivity of Arctic Black Carbon Radiative Effects to Subgrid Vertical Velocity in Aerosol Activation

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Abstract The representation of aerosol activation into cloud droplets in climate models is important for accurate understanding of aerosol radiative impacts on the Arctic climate, but it remains highly uncertain. Here we show that the uncertainty range of subgrid vertical velocity (SVV) and maximum supersaturation (SSmax) in aerosol activation produces fourfold to fivefold differences in the direct radiative effect of black carbon (BC) in the Arctic (0.091–0.40 W m⁻²) because SVV and SSmax determine the activated fraction and wet removal efficiency of aerosols. Aerosols are particularly sensitive to SVV in remote regions but not near their sources because many aerosols near sources are not yet influenced by wet removal processes. Our results demonstrate that SVV treatment is a major source of uncertainty in Arctic aerosol simulations and may be key for reducing the large discrepancies among global models in estimates of BC and its radiative effects in the Arctic.

Plain Language Summary Black carbon aerosol, emitted mainly in midlatitude regions by combustion of fossil fuel and biomass, is transported to the Arctic and deposited on snow and ice surfaces, where it contributes to Arctic heating. However, estimates of its importance in Arctic warming have large uncertainties. Because global climate models usually use a coarse horizontal grid spacing, they rely on many assumptions to represent the small-scale atmospheric processes within clouds. This study uses a global climate model to investigate the importance of one of these assumptions, the treatment of “subgrid vertical velocity,” to aerosol simulations. We show that varying the subgrid vertical velocity within its uncertainty range changes the calculated heating effect of black carbon in the Arctic by as much as five times. Our results underscore the importance of treating subgrid vertical velocity treatment accurately in estimating how much black carbon from midlatitudes is warming the Arctic.

1. Introduction

Black carbon (BC) aerosol, which is emitted from the incomplete combustion of fossil fuel, biofuel, and biomass, is considered to have a large positive direct radiative forcing through its absorption of solar radiation (e.g., Bond et al., 2013; Jacobson, 2001). However, estimates of this forcing are highly uncertain (e.g., Bond et al., 2013; Myhre, Samset, et al., 2013), and it is an important source of uncertainty in radiative forcing from all aerosol species and hence in human-induced climate change (Boucher et al., 2013; Myhre, Shindell, et al., 2013). BC-containing particles also modify cloud physical properties and precipitation by changing vertical atmospheric stability through absorption of solar radiation and by acting as cloud condensation nuclei (e.g., Koch & Del Genio, 2010; Stjern et al., 2017). BC particles deposited on snow and ice surfaces decrease surface albedo and lead to warming and melting (e.g., Clarke & Noone, 1985; Flanner, 2013). This BC snow albedo effect may have a large radiative forcing regionally (e.g., Flanner et al., 2007) and may enhance snow-albedo feedback in the Arctic (Serreze & Barry, 2011).

Although local and high-latitude emissions are important sources of Arctic BC (e.g., Huang et al., 2015; Stohl et al., 2013), large amounts of BC emitted in midlatitude regions (East Asia, Europe, and North America) are also considered to be transported to the Arctic, particularly during winter and early spring (e.g., Ikeda et al., 2017; Liu et al., 2015; Wang et al., 2011). BC particles of midlatitude origin are influenced during transport to the Arctic by atmospheric aging processes, which enhance their hygroscopicity through condensation, coagulation, and oxidation, and by their removal from the atmosphere through dry and wet deposition. Given the large uncertainties in the representation of these aging and removal processes in aerosol models, multimodel intercomparison studies have shown that simulated BC concentrations over the
Arctic vary by more than 2 orders of magnitude between models (e.g., Eckhardt et al., 2015; Koch et al., 2009). Reducing such uncertainties is essential to improve our understanding of the radiative impact of BC on the Arctic climate.

BC particles are activated as cloud condensation nuclei when the maximum supersaturation (SSmax) in clouds (determined mainly by cloud physics) is greater than the critical supersaturation (SScrit) of BC particles (determined by aerosol properties) (Seinfeld & Pandis, 2006). The representation of both SSmax and SScrit has large uncertainties in current global aerosol models. One cause of the large uncertainties in Arctic BC simulations is the treatment of hygroscopicity (or SScrit) for BC-containing particles. Most aerosol models have used a single threshold between hydrophobic (fresh, assumed not to be activated as cloud droplets) and hydrophilic (aged, assumed to be activated as cloud droplets) BC particles, and some studies have shown that Arctic BC concentrations are highly sensitive to this threshold and the representation of aging processes (e.g., Liu et al., 2011, 2016; Matsui & Mahowald, 2017; Matsui et al., 2018). However, because SSmax depends on cloud types and aerosol concentrations, using a single threshold to define hydrophobic and hydrophilic BC is problematic (Matsui, 2016). Moreover, absent a clear definition, the threshold can be assumed subjectively as a tuning parameter that causes simulations to agree well with observations.

Another potential cause of the large uncertainties in Arctic BC simulations, less often considered, is the treatment of the subgrid vertical velocity (SVV) in aerosol activation for grid-resolved clouds. Global model simulations, which usually use a coarse horizontal grid spacing (~100 km), cannot resolve fine structures of cloud-scale motions. Thus, the subgrid variability of vertical velocity is parameterized as a characteristic vertical velocity or a probability density function of vertical velocities (e.g., Golaz et al., 2011; Morrison & Gettelman, 2008; Wang & Penner, 2009). This treatment of SVV has a large uncertainty, yet it strongly influences SSmax, microphysics, and radiative effect of clouds through aerosol activation (e.g., Golaz et al., 2011; Tonttila et al., 2015; West et al., 2014). However, the importance of SVV treatment to aerosol distributions has not been closely evaluated.

Here we investigate the importance of SVV treatment in activation to aerosol concentrations and their radiative effects by using a global aerosol model that resolves particle size and BC mixing state. We find that the atmospheric burden and radiative effect of BC are not sensitive to SVV treatment over near-source regions but are highly sensitive to SVV treatment in the Arctic. We show that the current uncertainty range of SVV produces up to fivefold differences in BC burden, BC direct radiative effect, and total aerosol direct radiative effect in the Arctic, making SVV treatment one of the largest uncertainties in aerosol simulations for the Arctic.

2. Methods

The Community Atmosphere Model version 5 (CAM5) (Lamarque et al., 2012; Neale et al., 2012) with the Aerosol Two-dimensional bin module for foRmation and Aging Simulation (CAM5-chem/ATRAS) (Matsui, 2017; Matsui & Mahowald, 2017) considers the following atmospheric chemistry and aerosol processes: emissions, gas-phase chemistry (Emmons et al., 2010), nucleation (Kulmala et al., 2006; Matsui, Kojie, et al., 2011), condensation of sulfate, nitrate, and organic aerosols (Matsui, Kojie, Kondo, Fast, et al., 2014; Matsui, Kojie, Kondo, Takami, et al., 2014; Matsui, 2017), coagulation (Jacobson et al., 1994; Matsui, 2017; Matsui et al., 2013), aerosol activation (Abdul-Razzak & Ghan, 2000), aqueous-phase chemistry (Tie et al., 2001), dry deposition (Wesely, 1989; Zender et al., 2003), wet deposition (Liu et al., 2012; Rasch et al., 2000), aerosol-radiation interactions (Iacono et al., 2000), and aerosol-cloud interactions (Gettelman et al., 2010; Morrison & Gettelman, 2008). Aerosol particles from 1 nm to 10 μm in dry diameter are resolved with 12 size bins, and BC mixing state for fine particles (40 nm to 1.25 μm) in each size bin is resolved with eight bins, representing pure BC and BC-free particles plus six categories of internally mixed BC particles. The hygroscopicity (SScrit) and optical properties of aerosol particles are calculated based on the κ-Köhler theory (Petters & Kreidenweis, 2007) and the Mie theory (shell-core treatment) (Bohren & Huffman, 1998) for each size and mixing state bin (Matsui, 2017). As the CAM5-chem/ATRAS model can calculate particle-sized and mixing-state-dependent SScrit theoretically, it does not require a single assumed threshold between hydrophobic and hydrophilic BC. Aerosol particles in each two-dimensional bin are activated whenever SSmax is greater than SScrit (for grid-resolved clouds).
CAM5 uses the characteristic vertical velocity method to estimate SVV as Equation 1 (Neale et al., 2012):

$$\tilde{w} = \max \left( \bar{w} + c \sqrt{TKE}, w_{\text{min}} \right)$$

where $\tilde{w}$ is the characteristic vertical velocity, $\bar{w}$ is the mean vertical velocity in a grid box, $w_{\text{min}}$ is the minimum value of $\tilde{w}$, $c$ is a constant factor, and $TKE$ is turbulence kinetic energy. CAM5 originally set the values of $c$ at 0.82 ($\sqrt{2/3}$) and $w_{\text{min}}$ at 0.20 m s$^{-1}$, and we used these values in the Base simulation (supporting information Table S1). Considering previous studies (supporting information Text S1), we defined the uncertainty ranges of $c$ and $w_{\text{min}}$ as 0.10–1.0 and 0.1–0.4 m s$^{-1}$, respectively, and made six sensitivity simulations with different $c$ and $w_{\text{min}}$ values within these uncertainty ranges (supporting information Table S1). Here we mainly use two sensitivity simulations to examine the sensitivity of aerosols to SVV: LowSVV with the minimum value of $\tilde{w}$ (based on $c = 0.10$ and $w_{\text{min}} = 0.10$ m s$^{-1}$) and HighSVV with the maximum value of $\tilde{w}$ (based on $c = 1.0$ and $w_{\text{min}} = 0.40$ m s$^{-1}$). These two simulations produce the lowest and highest values of SSmax, respectively, which in turn yield the smallest and largest amounts of aerosols activated as cloud droplets and removed from the atmosphere by precipitation. The range of SSmax in these simulations is also consistent with the current uncertainty range of observation-based SSmax estimates (see section 3). The other four sensitivity simulations were used to investigate the relative importance of $c$ and $w_{\text{min}}$. Their results show that the uncertainties in both $c$ and $w_{\text{min}}$ contribute almost equally to the high sensitivity of Arctic aerosols to SVV (supporting information Text S2). Because this study focuses on aerosol activation (uncertainties in SVV and SSmax), all aspects of the model other than SVV are the same for all simulations.

We emphasize that the CAM5-chem/ATRAS model can resolve changes in BC activation and removal rate caused by slight changes in SSmax (for grid-resolved clouds) because this model calculates size- and mixing state-dependent SScriv in detail. The impact of SVV on activation and removal processes and the resulting changes in global distributions of BC can be simulated explicitly in our model but cannot be represented properly in a model that considers only one or two BC mixing state categories or that does not resolve size distributions sufficiently.

Model simulations were performed for 6 years (2007–2012) with a horizontal resolution of 1.9° × 2.5° and 30 vertical layers. The last 5 years (2008–2012) were used for analysis. Meteorological fields were nudged by the Modern-Era Retrospective analysis for Research and Applications Version 2 (MERRA2) data for temperature and horizontal wind in the middle and upper troposphere (<800 hPa). We used the Coupled Model Intercomparison Project Phase 6 (CMIP6) emission data set for the year 2010 (Hoesly et al., 2018; van Marle et al., 2017) for all aerosol and precursor species other than dust and sea salt emissions, which were calculated online. The BC direct radiative effect (DRE) was calculated based on the method of Ghan (2013) as the difference in radiative flux at the top of the atmosphere between the online calculation that considers all aerosol species and a diagnostic calculation that excludes BC. The CAM5-chem/ATRAS model simulations have been evaluated previously for aerosol mass concentrations of BC, sulfate, nitrate, ammonium, dust, and organic aerosol, aerosol number concentrations (>10 nm in diameter), aerosol optical depth, and single scattering albedo by using available observations (Matsui & Mahowald, 2017). The simulated global-mean aerosol optical depth at the wavelength of 550 nm (0.13) was lower than the Moderate Resolution Imaging Spectroradiometer satellite value (0.16) but generally consistent with estimates by other global aerosol models (e.g., 0.12 ± 0.025 in Kinne et al. (2006)). Comparisons between ATRAS and the Modal Aerosol Model, the default aerosol model in CAM, have shown that ATRAS yields slightly higher BC mass concentrations, aerosol optical depths, and cloud condensation nuclei concentrations (Matsui & Mahowald, 2017; supporting information Text S3).

We used BC mass concentrations observed at two surface sites, Barrow (71.3°N, 156.6°W) and Ny-Ålesund (78.9°N, 11.9°E), in the Arctic (Sinha et al., 2017) and during some aircraft campaigns over East Asia, Pacific, and Arctic (Kondo et al., 2011; Matsui, Kondo, et al., 2011; Oshima et al., 2012; Schwarz et al., 2010). We also used observation-based SSmax values estimated at midlatitude (Japan) (Moteki et al., 2019) and in the Arctic (Koike et al., 2019; Leaitch et al., 2016) to evaluate our model simulations (supporting information Text S4).
3. Results

Simulated SSmax values are within the range of observation-based estimates of SSmax in Japan and the Arctic (see detailed descriptions in supporting information Text S4). Although SSmax in the Base simulation tends to be higher than the observations in Japan and lower than the observations in the Arctic, the range of SSmax between the LowSVV and HighSVV simulations is comparable to its range in the observation-based estimates for both regions. These results suggest that the range of SSmax in our simulations is representative of the current uncertainty range of observation-based SSmax estimates.

Our Base simulation underestimates the observed surface BC mass concentrations (by 80–85%) at Barrow and Ny-Ålesund in winter and early spring (November–April) but reproduces them reasonably well in summer (supporting information Figure S1). Nevertheless, another study using our model has shown that simulated BC mass concentrations in precipitation and BC deposition flux are generally consistent with observations throughout the year (Mori et al., 2020). Therefore, the model may simulate BC mass in the middle and low troposphere, where clouds exist, reasonably well but may not simulate the vertical transport and distribution of BC well. BC emissions in the Arctic are highly uncertain (Huang et al., 2015; Stohl et al., 2013), which may also contribute to the underestimation of BC near the surface. The reduction of BC wet removal in mixed-phase clouds due to the Wegener-Bergeron-Findeisen process, which is not considered in our model, may be another reason for the underestimation of BC in our model (Ding et al., 2019; Qi et al., 2017). Qi et al. (2017) estimated that this process can result in a 25–70% increase in surface BC mass concentrations in the Arctic during winter and early spring. In addition, meteorological fields may contribute to the uncertainty in BC simulations because the observation and simulation years are not exactly the same and the horizontal grid resolution of our simulations (1.9° × 2.5°) is relatively coarse.

The HighSVV simulation underestimates BC mass concentrations even more strongly than the Base case (by 85–90%) at both Arctic sites in winter and early spring (supporting information Figure S1). The LowSVV simulation produces a better agreement with observations, underestimating BC mass concentrations by 40% at Barrow and overestimating them by 45% at Ny-Ålesund in these months. The ranges of BC mass concentrations in the SVV sensitivity simulations are especially large in these months (4.2–15 ng m⁻³ at Barrow and 3.5–34 ng m⁻³ at Ny-Ålesund). The ratio of these BC mass concentrations (LowSVV/HighSVV) is 3.7 at Barrow and 9.9 at Ny-Ålesund in winter and early spring (and 2.2 at Barrow and 3.4 at Ny-Ålesund in summer), showing that BC surface concentrations in the Arctic are highly sensitive to SVV treatment.

BC concentrations are sensitive to SVV not only at the surface but also throughout the troposphere (supporting information Figure S2). The LowSVV simulation yields BC mass concentrations that are closer to observations at middle and high latitudes in the northern hemisphere but deviate more from observations at low latitudes in the upper troposphere. The HighSVV simulation yields BC mass concentrations that are closer to observations in the upper troposphere at low latitudes but are more strongly underestimated at the surface in the Arctic. The SVV treatment cannot simultaneously improve the agreement with observations in the Arctic lower troposphere and in the tropical upper troposphere. However, the LowSVV/HighSVV ratios of BC mass concentration, 2–3 at low and middle latitudes and about 5 in the Arctic, show that BC mass concentrations are highly sensitive to SVV throughout the troposphere, especially in the Arctic. SVV treatment therefore may be one of the largest sources of uncertainty in BC simulations in the Arctic. SVV and its influence on SSmax thus may be an additional determinant of BC concentrations beyond those (hygroscopicity, deposition velocity on snow/ice surfaces, and aging processes) considered in previous studies.

Previous studies have shown that enhanced BC removal (especially by convective clouds) can simulate BC mass concentrations consistent with aircraft observations in the upper troposphere (He et al., 2016; Lund et al., 2018; Samset et al., 2014; Wang, Heald et al., 2014; Wang, Jacob et al., 2014; Yu et al., 2019). For example, Yu et al. (2019) showed that a modified convective transport scheme, which considers secondary activation from entrained air above the cloud base, reduces BC concentrations in the upper troposphere at low latitudes by 2 orders of magnitude. This process, not considered in our model, may explain why our simulations overestimate BC in the upper troposphere at low latitudes. These studies have suggested a typical BC lifetime of 3–4 days, which is shorter than the multimodel mean of 7.3 ± 2.3 days in the AeroCom phase I experiment (Schulz et al., 2006). The range of global BC lifetime is 4.4 (HighSVV) to 6.1 days (LowSVV) in our sensitivity simulations and is within the range of these previous estimates.
The global aerosol burden is not strongly sensitive to SVV treatment, differing by less than 10% for all non-BC aerosol species and by 38% for BC between the LowSVV and HighSVV simulations (Figure 1a). In contrast, the aerosol burden in the Arctic (>70°N) is much more sensitive to SVV treatment. The BC burden is 5 times greater in the LowSVV simulation than in the HighSVV simulation, and the sulfate, nitrate, ammonium, dust, and organic aerosol burdens differ by 60% to 140% (Figure 1a). The LowSVV/HighSVV ratio for the BC burden is close to unity (<1.5) near source regions (over land) but much higher (>3) over remote oceans and at high latitudes, where aerosol concentrations are dominated by transport and removal processes (supporting information Figure S3). This result shows the importance of SVV treatment for estimates of long-range BC transport and removal processes, the resulting atmospheric concentrations, and their radiative effects in remote areas. The same pattern is true for non-BC species, but their LowSVV/HighSVV ratios are much lower than those for BC.

The contrast in the sensitivity of aerosols to SVV treatment between near-source and remote regions can be explained by differences in the efficiency and amounts of aerosol removal by precipitation. Because many aerosols near their source are not yet influenced by wet removal processes, aerosol concentrations near source regions do not differ greatly between the LowSVV and HighSVV simulations even though the LowSVV simulation incorporates a lower $w$, a lower fraction of activated aerosols, and less efficient wet removal than the HighSVV simulation. For example, Oshima et al. (2013) estimated that only ~20% of BC is removed by precipitation during transport from East Asian sources (mainly China) to the low troposphere over outflow regions (Yellow Sea and East China Sea). In contrast, aerosols over remote regions experience much more wet removal by cloud and precipitation processes during long-range transport. For example, Matsui, Kondo, et al. (2011) estimated that BC particles were almost entirely removed by precipitation (80–90% in spring and 99% in summer) during transport from Asia to the Arctic. Because the activated fraction and wet removal efficiency of aerosols (removal fraction per amount of precipitation) differ between the LowSVV and HighSVV simulations, the remaining atmospheric aerosols over remote regions are strongly dependent on SVV treatment (supporting information Figure S4). Thus, Arctic BC is sensitive not only to precipitation amounts during transport (as shown by previous studies) but also to the activated fraction and wet removal efficiency of aerosols, which are determined by the balance between SSmax (calculated from SVV) and $SS_{crit}$.

BC is more sensitive to SVV treatment than other aerosol species because it has a lower hygroscopicity ($SS_{crit}$) and is less easily activated as cloud droplets than non-BC species. The relationship between critical diameter (the threshold particle diameter for activation) and hygroscopicity based on the κ-Köhler theory shows that the critical diameter at hygroscopicity values of 0.1 and 0.01 (corresponding to thinly coated BC) has a range of 80–390 and 170–840 nm, respectively, when SSmax varies between 0.05% and 0.5% (supporting information Figure S5). Because atmospheric aerosols typically have volume median diameters of around 200 nm or larger (Seinfeld & Pandis, 2006), the activated fraction of BC particles...
strongly depends on both the degree of BC aging and SSmax. The activation of BC particles, therefore, is sensitive to the variation of SSmax in response to the SVV treatment. In contrast, highly hygroscopic species are much less sensitive to SVV treatment; for instance, the critical diameter of sulfate (hygroscopicity of 0.61 in our study) has a range of 50–200 nm when SSmax varies between 0.05% and 0.5% (supporting information Figure S5); thus, most sulfate aerosol (by mass) can activate as cloud droplets in both the LowSVV and HighSVV simulations within a realistic range of SSmax (0.05–0.5% in our model simulations; supporting information Figure S5). Dust, which has a low hygroscopicity (0.001) in our model, is relatively insensitive to SVV, likely because dry deposition, which is not related to SVV (or cloud and precipitation processes), accounts for about half of the global deposition flux of dust in our simulations. The sensitivity of dust to SVV may change depending on the choice of hygroscopicity and emission size distribution.

As with the case of BC burden, the sensitivity to SVV of BC surface concentrations, deposition flux, and DRE is low on the global scale (LowSVV/HighSVV = 1.0–1.4) but high in the Arctic (Figure 1b). The range of mean surface concentrations and deposition flux of BC in the Arctic is 4.3–17 ng m\(^{-3}\) (LowSVV/HighSVV = 3.9) and 39–77 Gg year\(^{-1}\) (LowSVV/HighSVV = 2.0), respectively, in the SVV sensitivity simulations. The range of BC DRE (at the top of the atmosphere) is 0.31–0.43 W m\(^{-2}\) (LowSVV/HighSVV = 1.4) globally and 0.091–0.40 W m\(^{-2}\) (LowSVV/HighSVV = 4.4) in the Arctic (Figure 2). The DRE for total aerosols in the Arctic (BC + non-BC) is also different by a factor of 5.3 (Figure 1b). These results demonstrate that BC and total aerosol radiative effects in the atmosphere and at the snow/ice surface in the Arctic are strongly influenced by SVV treatment.

BC deposition flux in the Arctic, which is mostly controlled by in-cloud scavenging in our simulations (Moteki et al., 2019), is less sensitive to SVV treatment (LowSVV/HighSVV = 2.0) than BC burden, surface concentrations, and DRE (LowSVV/HighSVV = 4.0–5.1) because smaller fractions of aerosol particles are activated as cloud droplets in the LowSVV simulation than in the HighSVV simulation due to lower SSmax. The lower SSmax in the LowSVV simulation leads to a smaller fraction of activated aerosols than in the HighSVV simulation (LowSVV/HighSVV = 0.73% globally and 0.36% in the Arctic) and contributes to weakening the sensitivity of BC deposition flux to SVV.

We note that our main conclusions in this section do not change even if meteorology, emissions, and the resulting simulations of aerosol distributions and removal processes and aerosol-cloud interactions are different (supporting information Figures S6 and S7).

We also emphasize that our Arctic simulations, which use eight BC mixing states, produce a much larger difference in BC burden between the LowSVV and HighSVV simulations (LowSVV/HighSVV = 5.1) than a model version with a single mixing state (LowSVV/HighSVV = 3.3) (Figure 1a). Because the multiple mixing state simulation can resolve pure BC and thinly coated BC particles, which have low hygroscopicity (high SScrit), BC activation is more sensitive to SVV treatment in our model. The single mixing state simulation, which assumes that all BC is internally mixed with other aerosol species, yields BC with uniformly high
hygroscopicity (low SScrit) that is less sensitive to SVV treatment. Similarly, we obtain larger differences in BC surface concentrations (LowSVV/HighSVV of 3.9 versus 2.0) and BC DRE (LowSVV/HighSVV of 4.4 versus 2.9) by resolving the BC mixing state (Figure 1b).

SVV treatment also changes aerosol number concentrations and their impact on clouds. Compared with the HighSVV simulation, the LowSVV simulation enhances number concentrations of aerosol particles greater than 40, 80, and 150 nm in dry diameter at the surface by 6.5%, 11%, and 12%, respectively, as global averages and by 100%, 169%, and 220%, respectively, in the Arctic. In contrast, column cloud droplet number concentrations are reduced by 73% globally and 77% in the Arctic due to the reduction of SVV. These results suggest that reduced (enhanced) SVV increases (decreases) aerosol concentrations and their radiative effects, especially over the Arctic and remote oceans, while also decreasing (increasing) the sensitivity of aerosol-cloud interactions on the global scale. Although our focus in this study is to understand the impact of SVV treatment on aerosol distributions, these results also suggest that modifications of SVV in future studies must be done by using evaluations of both aerosol and cloud fields that are based on available observations.

4. Discussion and Conclusions

Because modifying SVV values leads to radiatively unbalanced flux at the top of the atmosphere, mainly through aerosol-cloud interactions, other parameters in cloud microphysics must be tuned for radiatively balanced long-term climate simulations (e.g., Golaz et al., 2011). Golaz et al. (2011) pointed out that retuning of the autoconversion threshold, which they did to resolve a radiatively unbalanced situation caused by an SVV modification, had a large impact on their estimates of radiative forcing by aerosol-cloud interactions from the preindustrial to present-day climate. The use of observations (e.g., the method of Moteki et al., 2019) or high-resolution model simulations (e.g., large-eddy simulations) to reduce the large uncertainties in the SVV treatment may be helpful not only to improve estimates of the long-range transport and radiative effects of aerosols but also to constrain other cloud microphysical parameters with large uncertainties that currently must be tuned in climate simulations.

This study used a particle-sized and mixing state-resolved global aerosol model to show that the current uncertainties in the treatment of SVV (only two parameters, c and wmin) change BC burden and DRE in the Arctic and over remote oceans by a factor of 4 or 5. BC burden and DRE are particularly sensitive to SVV treatment in the Arctic but not over near-source regions or on the global scale. BC DRE in the Arctic has a range of 0.091–0.40 W m⁻² when we change the SVV value within its current uncertainties. BC is more sensitive to SVV than other aerosol species because fresh (pure + thinly coated) BC has a lower hygroscopicity and higher SScrit and the activated fraction of BC-containing particles strongly depends on both the degree of BC aging (which changes SScrit) and SSmx. Our results demonstrate the importance of SSmx and SVV in estimates of the long-range transport of BC to the Arctic and the burden and radiative effects of BC in the Arctic, both in the atmosphere and on the snow/ice surface. Our conclusions also suggest that SVV, through its effect on estimation of SSmx, is a major source of uncertainty in aerosol simulations of the Arctic and may be a key parameter for reducing the large discrepancies in Arctic BC concentrations among current global aerosol models.

Conflict of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data used in Figures 1 and 2 are available from the link (http://doi.org/10.5281/zenodo.3960524). Other data can be obtained by contacting the corresponding author (H. M.).

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