Importance of Supersaturation in Arctic Black Carbon Simulations

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(Manuscript received 21 December 2020, in final form 7 June 2021)

ABSTRACT: Black carbon (BC) aerosol particles in the Arctic heat the atmosphere and snow/ice surfaces and may strengthen the snow-albedo feedback that amplifies Arctic warming. Model simulations of BC concentrations in the Arctic depend strongly on the representation of microphysical processes such as aging, activation, and wet removal. Most BC modeling studies have classified BC particles into hydrophobic BC, which cannot form cloud droplets, and hydrophilic BC, which can form cloud droplets, by assuming a globally constant critical supersaturation threshold value (S_{thre}), without considering its consistency with cloud maximum supersaturation (S_{max}). Here we show that it is essential to consider the consistency of S_{thre} with S_{max} in global model simulations to reduce uncertainties in near-surface ambient BC concentrations in the Arctic. Previous studies often obtained good agreement between simulated and observed near-surface Arctic BC mass concentrations when a low S_{thre} ($\sim 0.1\%$) was assumed in their models. However, this S_{thre} may be too low (activation and wet removal of BC may be underestimated) for the Arctic, because some recent observations and our model simulations suggest that S_{max} may actually be higher ($\sim 0.3\%$) there. We also demonstrate that spatially varying S_{thre} values and their consistency with S_{max} , which previous studies did not consider, must be represented in models for more accurate estimation of BC budget in the Arctic. Because both S_{max} and BC-aging speed depend on climatic conditions, our findings are an important step toward better simulations of BC impacts on past, present, and future Arctic climates.

KEYWORDS: Aerosols; Arctic; Aerosol-cloud interaction; Climate models; Global transport modeling; Atmospheric composition; Model evaluation/performance

1. Introduction

Black carbon (BC) aerosol particles are estimated to have a large positive radiative forcing through the absorption of solar radiation and the impact of this absorption on vertical atmospheric stability and cloud properties (aerosol–radiation interactions) (e.g., Bond et al. 2013; Boucher et al. 2013; Jacobson 2001; Myhre et al. 2013). In addition, BC deposited on surfaces of snow and ice decreases the surface albedo and contributes to their melting (snow-albedo effect) (e.g., Flanner et al. 2007; Hadley and Kirchstetter 2012). These heating effects of BC in the atmosphere and on snow and ice surfaces can have a large radiative forcing in the Arctic (e.g., Hansen and Nazarenko 2004; Matsui and Moteki 2020) and strengthen the snow-albedo feedback, which amplifies Arctic warming (e.g., Serreze and Barry 2011). However, simulations of BC in the Arctic are still highly uncertain because our understanding and model representation of BC microphysical processes such as aging, activation, and wet removal are inadequate. The resulting large discrepancies (of more than two orders of magnitude) in simulated BC concentrations over the Arctic among current global aerosol models (e.g., Eckhardt et al. 2015;

Koch et al. 2009; Sand et al. 2017) hinder accurate understanding of BC impacts on the Arctic climate and climate change.

BC particles in the atmosphere are gradually coated by non-BC species such as sulfate, nitrate, and organic material through condensation and coagulation processes (e.g., Moteki et al. 2007). These aging processes enhance the hygroscopicity and cloud condensation nuclei activity of BC and change its wet removal efficiency, lifetime, and global distribution (e.g., Stier et al. 2006). To represent these processes, previous modeling studies have developed various BC-aging parameterizations (e.g., Liu et al. 2011; Oshima and Koike 2013), which have been used in almost all BC modeling studies (e.g., He et al. 2016; Liu et al. 2016; Mahmood et al. 2016; Shen et al. 2017; Wang et al. 2014). Previous studies have shown that the agreement of modeling results with surface BC mass (M_{BC}) observations in the Arctic has been improved through the use of these parameterizations (Liu et al. 2011; Mahmood et al. 2016; Shen et al. 2017) and that M_{BC} concentrations in the Arctic are highly sensitive to the treatment of aging processes (e.g., Liu et al. 2016; Wang et al. 2014).

A subset of the aerosol particle population grows by water condensation in supersaturated clouds to become cloud droplets, which may be subsequently removed from the atmosphere by precipitation (e.g., Andreae and Rosenfeld 2008). Typically, the smallest size of particle that will become a cloud droplet depends on the particle's ability to grow beyond its critical diameter at the maximum supersaturation (S_{max}) achieved in the cloud, and that can be represented by the particle's critical supersaturation (S_{crit}) value, if we assume that the time needed to reach the critical diameter is sufficiently short (although some much larger particles may not have enough time to reach their critical diameter) (Abdul-Razzak and Ghan 2000;

Denotes content that is immediately available upon publication as open access.

Supplemental information related to this paper is available at the Journals Online website: <https://doi.org/10.1175/JCLI-D-20-0994.1.s1>.

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DOI: 10.1175/JCLI-D-20-0994.1

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FIG. 1. Schematic diagram showing relationships among the three supersaturation variables used in this study. $S_{\max}$ denotes the maximum supersaturation value in clouds, and S_{crit} denotes the critical supersaturation value determined by aerosol properties. The S_{crit} value of BC-containing particles decreases as aging progresses (arrows in the figure). S_{thre} is the threshold S_{crit} value between hydrophobic BC (which cannot form cloud droplets) and hydrophilic BC (which can form cloud droplets) defined in aerosol models. (a) A case when S_{thre} equals $S_{\max}$. BC-containing particles with S_{crit} lower than S_{thre} (red-shaded area) can be activated as cloud droplets. In this case, the calculated BC activation is theoretically correct. (b) A case when S_{thre} is lower than $S_{\max}$. BC-containing particles that have S_{crit} between S_{thre} and $S_{\max}$ (blue-shaded area) should be activated as cloud droplets, but their activation is not allowed in models. (c) A case when S_{thre} is higher than $S_{\max}$. BC-containing particles that have S_{crit} between $S_{\max}$ and S_{thre} (blue-shaded area) should not be activated as cloud droplets, but they are allowed to form cloud droplets in models.

Nenes and Seinfeld 2003). Under this assumption, all particles with S_{crit} equal to or smaller than $S_{\max}$ form cloud droplets.

Note that $S_{\max}$ can vary depending on the cloud type and the aerosol number size distribution (e.g., Abdul-Razzak and Ghan 2000), whereas S_{crit} depends strongly on the particle size, chemical composition, and mixing state of the aerosol particles (e.g., Petters and Kreidenweis 2007). Therefore, whether a BC-containing particle becomes a cloud droplet is determined by the relationship between $S_{\max}$ and S_{crit} , both of which can vary spatially and temporally. Aging processes increase the amount of coating and reduce the S_{crit} value of BC-containing particles. Thus, aged BC has a lower S_{crit} value than fresh BC and can form cloud droplets more easily. The threshold S_{crit} value between BC particles that can form cloud droplets (called hydrophilic BC in many studies) and BC particles that cannot form cloud droplets (called hydrophobic BC), denoted S_{thre} hereafter, is an important factor controlling the long-range transport, lifetime, distribution, and climate impacts of BC.

The values of S_{thre} and $S_{\max}$ should be equal in calculations of ambient BC particles to form cloud droplets (Fig. 1a). If $S_{\max}$ is high, then S_{thre} also needs to be high, and BC particles with a thin coating (high S_{crit} particles) are able to form cloud droplets and be wet scavenged by precipitation. Because in this case only a small amount of coating is required for the BC to form cloud droplets, the conversion rate of BC from hydrophobic to hydrophilic is fast. If $S_{\max}$ is low, S_{thre} should be low, and only BC particles with low S_{crit} values (aged BC) can be wet scavenged. In this case, a thicker coating is necessary for the particles to form cloud droplets; hence, the hydrophobic-to-hydrophilic BC conversion rate is relatively slow. In the developed BC-aging parameterizations (e.g., Liu et al. 2011; Oshima and Koike 2013), however, a globally constant S_{thre} has been used. Neither $S_{\max}$ nor the consistency between $S_{\max}$ and S_{thre} and their spatial/temporal variations is explicitly considered in these BC-aging parameterizations. For this reason, the S_{thre} value assumed in these parameterizations is not necessarily consistent

with $S_{\max}$ in clouds. When $S_{\max}$ is greater than the assumed S_{thre} (Fig. 1b), BC-containing particles that have S_{crit} between S_{thre} and $S_{\max}$ (blue-shaded region in Fig. 1b) should form cloud droplets, but when these previous parameterizations are used, such BC-containing particles do not form cloud droplets in model simulations. When $S_{\max}$ is lower than S_{thre} (Fig. 1c), BC-containing particles that have S_{crit} between $S_{\max}$ and S_{thre} (blue-shaded region in Fig. 1c) cannot form cloud droplets, but with these previous parameterizations, they are allowed to form cloud droplets in model simulations.

Here we show that it is necessary to consider the consistency between $S_{\max}$ and S_{thre} in global climate models to reduce the uncertainties in Arctic BC simulations. Our results indicate that the inconsistency between $S_{\max}$ and S_{thre} in the Arctic leads to theoretically incorrect BC activation and incorrect understanding of the Arctic BC budget even if the model achieves good agreement between simulated results and observed surface M_{BC} in the Arctic. We demonstrate that spatial variations of $S_{\max}$ and S_{thre} and their consistency, which are not considered properly by current BC-aging parameterizations, should be represented by using a mixing-state-resolved model or a parallel aging parameterization to obtain more accurate Arctic BC simulations in the past, present, and future climate conditions.

2. Methods

a. Global model simulations

Global BC simulations were conducted by using the Community Atmosphere Model version 5 (Lamarque et al. 2012; Liu et al. 2012) with the Aerosol Two-Dimensional Bin Module for Formation and Aging Simulation (CAM5-chem/ATRAS) (Matsui 2017, 2020; Matsui and Mahowald 2017; Matsui et al. 2018a,b). Aerosol processes considered in the CAM5-chem/ATRAS model are nucleation; condensation of sulfate, ammonium, and nitrate;

TABLE 1. Summary of the model simulations. Group 1 simulations assume a faster hydrophobic-to-hydrophilic BC conversion rate and tend to underestimate observed near-surface M_{BC} in the Arctic. Group 2 simulations assume a slower hydrophobic-to-hydrophilic BC conversion rate and show better agreement with near-surface M_{BC} observations in the Arctic.

Run	BC aging	S_{thre}		Group
		<60°N	>60°N	
Base	MS-resolved	S_{max} (~0.15%)	S_{max} (>0.3%)	1
$S = 0.1\%$ (global)	MS-resolved	<0.1%	<0.1%	2
$S = 0.1\%$ (>60°N)	MS-resolved	S_{max} (~0.15%)	<0.1%	2
$S = 0.3\%$ (>60°N)	MS-resolved	S_{max} (~0.15%)	<0.3%	—
$Emis \times 2$ (>60°N)	MS-resolved	S_{max} (~0.15%)	S_{max} (>0.3%)	—
Param_0.3%	Parameterization	0.3%	0.3%	1
Param_0.1%–0.3%	Parameterization	0.1%	0.3%	1
Param_0.1%	Parameterization	0.1%	0.1%	2
Param_Liu2011	Parameterization	^a	^a	2
Param_24h	Parameterization	^b	^b	1

^a The hydrophobic-to-hydrophilic BC conversion rate was assumed to be $\beta[OH] + \delta$, where β ($= 4.6 \times 10^{-12}$ cm³ per molecule per second) and δ ($= 5.8 \times 10^{-7}$ s^{−1}) are constants for fast (condensation) and slow (coagulation) aging processes, and $[OH]$ is the hydroxyl radical concentration.

^b The hydrophobic-to-hydrophilic BC conversion rate was assumed to be the reciprocal of 24 h.

secondary organic aerosol formation; aerosol activation; aqueous-phase chemistry; dry and wet deposition; aerosol–radiation interactions; and aerosol–cloud interactions (Matsui 2017; Matsui and Mahowald 2017). Aerosol particles are represented by using a two-dimensional sectional method in which dry particle diameters from 1 to 10 000 nm are resolved into 12 bins, and the BC mixing state into 8 bins. The eight BC mixing state bins are defined by the fraction of BC mass relative to the total dry aerosol mass (f_{BC}): BC-free particles ($f_{BC} < 0.0001$), six categories of internally mixed BC particles ($f_{BC} = 0.0001$ –0.1, 0.1–0.2, 0.2–0.5, 0.5–0.8, 0.8–0.9, and 0.9–0.99), and pure BC particles ($f_{BC} = 0.99$ –1.0). The S_{crit} value is calculated for the individual two-dimensional bins (Matsui 2016) by using κ –Köhler theory (Petters and Kreidenweis 2007). The S_{max} value is calculated as a function of updraft velocity and aerosol number concentrations in the aerosol activation scheme for liquid-phase clouds (Abdul-Razzak and Ghan 2000). Aerosol particles in each two-dimensional bin can form cloud droplets whenever S_{max} is greater than S_{crit} . Cloud-phase and interstitial aerosols are represented separately in CAM5-chem/ATRAS, as in the original CAM5 (Ghan and Easter 2006; Liu et al. 2012). The CAM5-chem/ATRAS model does not need to assume a single threshold between hydrophobic and hydrophilic BC, as is done in most global aerosol models, because the relationship between S_{max} and S_{crit} is calculated explicitly for each particle size and mixing state bin. Our recent study also substantially improved Arctic BC simulations by modifying wet deposition processes of aerosols for mixed-phase clouds and convective clouds (Liu and Matsui 2021b). To our knowledge, no other global aerosol models consider particle-size- and mixing-state-dependent aging processes and the S_{max} – S_{crit} relationship with a spatially and temporally variable S_{thre} , while they are treated explicitly in some box models (e.g., Riemer et al. 2009).

CAM5-chem/ATRAS simulations have been evaluated against observations in our previous studies for aerosol mass concentrations of BC, organic aerosols, sulfate, nitrate, and dust, aerosol number concentrations, aerosol optical properties, and

BC mixing state (Liu and Matsui 2021a; Matsui and Mahowald 2017; Matsui et al. 2014, 2013). Precipitation amounts in the Arctic have also been simulated well at two observation sites in the Arctic: Barrow, Alaska (71.3°N, 156.6°W; now known as Utqiagvik), and Ny-Ålesund, Norway (78.9°N, 11.9°E) (Mori et al. 2020).

All model simulations in this study were performed for the 6 years between 2007 and 2012 with a resolution of $1.9^\circ \times 2.5^\circ$ horizontally and 30 levels vertically. The first year was used for spinup, and the remaining five years (between 2008 and 2012) were used for analysis. Meteorological nudging was done by using Modern-Era Retrospective Analysis for Research and Applications version 2 (MERRA-2) data for temperature and horizontal wind in the free troposphere (<800 hPa). We used anthropogenic and biomass burning emissions for phase 6 of the Coupled Model Intercomparison Project (CMIP6; Hoesly et al. 2018; van Marle et al. 2017) for the year 2010.

We conducted nine model simulations, each of which used a different treatment for BC aging, activation, and removal processes (Table 1). The Base simulation used the standard CAM5-chem/ATRAS setup and explicitly calculated the relationship between S_{max} and S_{crit} with the particle-size- and BC-mixing-state-resolved representation. Three sensitivity simulations, $S = 0.1\%$ (global), $S = 0.1\%$ (>60°N), and $Emis \times 2$ (>60°N), also used the particle-size- and BC-mixing-state-resolved representation. In $S = 0.1\%$ (global) and $S = 0.1\%$ (>60°N), we changed the in-cloud aerosol scavenging scheme so that aerosol particles with S_{crit} greater than 0.1% were not removed by this process, globally and at high latitudes (>60°N), respectively. In these simulations, even if aerosol particles with $S_{crit} > 0.1\%$ can form cloud droplets (when S_{max} is greater than 0.1%), they are not removed from the atmosphere through in-cloud scavenging processes; instead, in-cloud aerosol scavenging occurs only for particles with $S_{crit} < 0.1\%$. This treatment is generally similar to a treatment that assumes the threshold of S_{crit} between hydrophobic and hydrophilic BC (S_{thre}) to be less than 0.1%. In $Emis \times 2$ (>60°N),

BC emission fluxes at high latitudes ($>60^\circ\text{N}$) were doubled to examine the sensitivity of near-surface M_{BC} in the Arctic to BC emission fluxes at high latitudes (section 3c). In other respects, the setups of the three sensitivity simulations are the same as the Base simulation setup.

For the other five sensitivity simulations, we used BC-aging parameterizations (Table 1). The single-mixing-state version of CAM5-chem/ATRAS (Matsui and Mahowald 2017) was used in these simulations. Particle size was resolved into 12 size bins by assuming internally mixed particles in each size bin. In Param_Liu2011 and Param_24h, the hydrophobic BC mass ($M_{\text{BC,pho}}$) at each time step was calculated with Eq. (1):

$$M_{\text{BC,pho},t+\Delta t} = M_{\text{BC,pho},t} \exp(-k\Delta t), \quad (1)$$

where Δt is the time step of the model simulation (30 min) and k is the hydrophobic-to-hydrophilic conversion rate of M_{BC} . This equation is considered in most BC-aging parameterizations (e.g., Liu et al. 2011; Oshima and Koike 2013; Vignati et al. 2010). The conversion rate k for Param_Liu2011 is obtained with Eq. (2):

$$k = \beta[\text{OH}] + \delta, \quad (2)$$

where β ($=4.6 \times 10^{-12} \text{ cm}^3$ per molecule per second) and δ ($=5.8 \times 10^{-7} \text{ s}^{-1}$) are constants for fast (condensation) and slow (coagulation) aging processes, and $[\text{OH}]$ is the hydroxyl radical concentration (Liu et al. 2011). The conversion rate k for Param_24h is $1.2 \times 10^{-5} \text{ s}^{-1}$ (the inverse of 24 h). In Param_0.3% and Param_0.1%, $M_{\text{BC,pho}}$ was calculated with Eq. (3):

$$M_{\text{BC,pho},t+\Delta t} = M_{\text{BC,pho},t} \left(1 - \frac{ab}{C_\kappa^S} \Delta t \right) \exp(-\delta\Delta t), \quad (3)$$

where a is the volume condensation rate of secondary aerosols (sulfate, ammonium, nitrate, and secondary organic aerosols) at each time step, b is the volume fraction of hydrophobic BC to total aerosols, and C_κ^S is the volume of secondary aerosols required to convert all hydrophobic BC to hydrophilic BC. The terms C_κ^S , a , and b are calculated for the bulk aerosol (sum of all particle-size bins). The hydrophobic-to-hydrophilic conversion fraction at a time step ($1 - M_{\text{BC,pho},t+\Delta t}/M_{\text{BC,pho},t}$) is therefore assumed to be the same for all bins in these calculations. Equation (3) is similar to Eqs. (1) and (2), but in Eq. (2), BC aging by condensation processes is represented as a function of $[\text{OH}]$, whereas in Eq. (3) it is represented by explicit calculations of the condensation rate. The term C_κ^S depends on S_{thre} and the hygroscopicity (κ) of the secondary aerosols produced by condensation at each time step and was calculated by assuming that the dry diameter of hydrophobic BC is 150 nm. This value was chosen because previous BC observations made with a single-particle soot photometer (SP2) have shown the mass median diameter of BC to be 150–200 nm in both the near-source and remote atmosphere, but 150–300 nm in the Arctic (Moteki et al. 2012; Schwarz et al. 2010; Yoshida et al. 2020). The hydrophobic/hydrophilic partitioning in Eq. (3) affects nucleation scavenging only; water uptake is calculated by the κ -Köhler theory using the information on aerosol

composition and particle size in each size bin. In Param_0.1%–0.3%, two S_{thre} values (0.1% and 0.3%) were used simultaneously to calculate hydrophobic BC:

$$M_{\text{BC,pho},t+\Delta t}^{S=0.1\%} = \min \left[M_{\text{BC,pho},t}^{S=0.1\%} \left(1 - \frac{ab}{C_\kappa^{S=0.1\%}} \Delta t \right) \times \exp(-\delta\Delta t), M_{\text{BC,tot},t+\Delta t} \right] \quad (4a)$$

$$M_{\text{BC,pho},t+\Delta t}^{S=0.3\%} = \min \left[M_{\text{BC,pho},t}^{S=0.3\%} \left(1 - \frac{ab}{C_\kappa^{S=0.3\%}} \Delta t \right) \times \exp(-\delta\Delta t), M_{\text{BC,tot},t+\Delta t} \right] \quad (4b)$$

where $M_{\text{BC,pho}}^{S=0.1\%}$ and $M_{\text{BC,pho}}^{S=0.3\%}$ are the hydrophobic M_{BC} concentrations when S_{thre} is 0.1% and 0.3%, respectively, and $M_{\text{BC,tot}}$ is the total (hydrophobic + hydrophilic) M_{BC} concentration. The latitudinal dependence of S_{thre} is considered by using Eqs. (4a) and (4b) simultaneously: S_{thre} at 0.1% was used in low and middle latitudes ($<60^\circ\text{N}$), and S_{thre} at 0.3% was used in high latitudes ($>60^\circ\text{N}$).

We also conducted additional sensitivity simulations for the Base and $S = 0.3\%$ ($>60^\circ\text{N}$) cases to explore the sensitivity of the BC aging and S_{max} effects to emission reductions (section 3d). The treatment of $S = 0.3\%$ ($>60^\circ\text{N}$) is similar to that of $S = 0.1\%$ ($>60^\circ\text{N}$) except for the S_{thre} value: $S = 0.3\%$ ($>60^\circ\text{N}$) assumes S_{thre} to be less than 0.3% at high latitudes ($>60^\circ\text{N}$). In these sensitivity simulations, anthropogenic and biomass burning emissions of all aerosols and their precursor species except dust and sea salt were reduced to 10%, 20%, or 50% of present-day emissions globally. Simulations for a preindustrial condition using the CMIP6 emissions (for year 1750) and for a future condition using the RCP4.5 emissions (Moss et al. 2010) were also performed for the Base and $S = 0.3\%$ ($>60^\circ\text{N}$) cases.

b. Hydrophilic M_{BC} fraction

The fraction of hydrophilic M_{BC} relative to total (hydrophobic + hydrophilic) M_{BC} was used to calculate BC statistics. In the simulations using BC-aging parameterizations, the hydrophilic M_{BC} fraction was calculated as $1 - M_{\text{BC,pho}}/M_{\text{BC,tot}}$. In the Param_0.1%–0.3% simulation, the fraction was calculated by using $M_{\text{BC,pho}}$ at $S_{\text{thre}} = 0.3\%$. In the particle-size- and BC-mixing-state-resolved simulations, the hydrophilic M_{BC} fraction was calculated by assuming $S_{\text{thre}} = 0.3\%$ in Base and Emis $\times 2$ ($>60^\circ\text{N}$), and $S_{\text{thre}} = 0.1\%$ in $S = 0.1\%$ (global) and $S = 0.1\%$ ($>60^\circ\text{N}$).

c. Observation data for model evaluations

Surface M_{BC} concentrations at Barrow and Ny-Ålesund (Sinha et al. 2017) were used for evaluating the monthly variations of simulated surface M_{BC} in the Arctic. We also used M_{BC} concentrations obtained during the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observations (HIPPO) research aircraft campaigns (Schwarz et al. 2013) for evaluating the vertical

FIG. 2. Global distribution of cloud maximum supersaturation ($S_{\max}$) in the Base simulation. $S_{\max}$ values are averaged in the lower troposphere (>800 hPa) only over layers and periods in which clouds exist.

profiles of simulated M_{BC} at high latitudes in the Northern Hemisphere ($>60^\circ\text{N}$).

3. Results

a. $S_{\max}$ and S_{thre}

The global distribution of $S_{\max}$ in the Base simulation (Table 1) shows a clear latitudinal dependence: higher $S_{\max}$ ($>0.3\%$) at high latitudes ($>60^\circ\text{N}$) and lower $S_{\max}$ ($\sim 0.1\%$) at low and middle latitudes (Fig. 2). This latitudinal dependence is generally consistent with the results of a previous study that simulated $S_{\max}$ global distributions (Pringle et al. 2009) and is obtained because concentrations of aerosols, which contribute to the reduction of $S_{\max}$ by water uptake in supersaturated clouds, are low at high latitudes. In our Base simulation, BC particles with low S_{crit} ($<0.1\%$, to the right of the red line in Fig. 3) are activated at all latitudes, whereas BC particles with higher S_{crit} ($0.1\%–0.3\%$, particles between the light blue and red lines in Fig. 3) are activated only at high latitudes, where $S_{\max}$ is higher. This difference in BC activation (i.e., the activated BC fraction) caused by the $S_{\max}$ latitudinal dependence can be calculated explicitly in our particle-size- and mixing-state-resolved model but cannot be treated properly in models using BC-aging parameterizations.

In addition to the Base simulation, we conducted eight sensitivity simulations in which aging, activation, and wet removal processes were treated differently (Table 1). Five of the nine simulations—Base, Emis $\times 2$ ($>60^\circ\text{N}$), Param_0.3%, Param_0.1%–0.3%, and Param_24h—used a high S_{thre} value ($\sim 0.3\%$) (the hydrophobic-to-hydrophilic conversion rate is faster) in the Arctic; we call these simulations, except Emis $\times 2$ ($>60^\circ\text{N}$), Group 1 hereafter (Table 1). The other four simulations— $S = 0.1\%$ (global), $S = 0.1\%$ ($>60^\circ\text{N}$), Param_0.1%, and Param_Liu2011—used a low S_{thre} value ($\sim 0.1\%$) (the hydrophobic-to-hydrophilic conversion rate is slower) in the Arctic, and we call these simulations Group 2 hereafter. Compared with Group 2, Group 1 simulates larger BC activation and wet removal amounts and lower atmospheric M_{BC} concentrations in the Arctic.

FIG. 3. Particle-size and BC-mixing-state dependence of critical supersaturation (S_{crit}). $S_{\text{crit}} = 0.3\%$ (light blue) and $S_{\text{crit}} = 0.1\%$ (red) lines were calculated based on κ -Köhler theory by assuming a mixture of BC ($\kappa = 10^{-6}$) and ammonium sulfate ($\kappa = 0.61$). BC-containing particles on the right side of the $S_{\text{crit}} = 0.3\%$ and $S_{\text{crit}} = 0.1\%$ lines (larger particle sizes and smaller BC mass fractions) can activate as cloud condensation nuclei when cloud maximum supersaturation ($S_{\max}$) is 0.3% and 0.1% , respectively. Thin gray lines show the particle-size and BC-mixing-state bins used in our global aerosol model. In our model, there are 12 particle-size bins between 1 and 10 000 nm in dry diameter and 8 BC-mixing-state bins. BC-mixing-state bins are defined by the fraction of BC to total dry aerosol mass for fine particles (between 40 and 1250 nm in dry diameter) (Matsui and Mahowald 2017).

b. Relationship between Arctic M_{BC} and S_{thre}

Near-surface M_{BC} in the Arctic shows distinct seasonal variations: M_{BC} is higher in winter/spring and lower in summer (Figs. 4a,b). In winter and spring, liquid (supercooled), ice, and mixed-phase clouds exist in the Arctic. BC emitted at high latitudes (e.g., northern Siberia) within the Arctic dome is transported at lower altitudes. BC emitted at middle latitudes is transported at higher altitudes and through a long distance to the Arctic, but the air parcels may subside in the Arctic (e.g., AMAP 2015; Stohl 2006). Although wet removal by precipitation is considered to be slow in the Arctic (e.g., Shen et al. 2017), BC transport from the midlatitudes to the Arctic depends strongly on activation and wet removal processes during the long-range transport (e.g., Matsui and Moteki 2020). In summer, liquid-phase clouds are dominant and the precipitation rate is much higher in the Arctic, leading to efficient wet removal of BC particles (e.g., Shen et al. 2017). In this season, BC from biomass burning is considered to be more important in the Arctic (e.g., Winiger et al. 2016, 2017).

Group 1 (BASE simulation) severely underestimates the observed surface M_{BC} concentrations in the Arctic during winter (December–February) and early spring, but Group 2 [$S = 0.1\%$ (global) and $S = 0.1\%$ ($>60^\circ\text{N}$)] simulates surface M_{BC} closer to observations and reproduces seasonal variations reasonably well, with maximum M_{BC} in winter (Figs. 4a,b; see also Fig. S1 in the online supplemental material). These results are consistent with the findings of previous studies (Liu et al. 2011,

FIG. 4. Observed and simulated BC mass (M_{BC}) concentrations in the Arctic. Monthly variations of surface M_{BC} concentrations at (a) Barrow and (b) Ny-Ålesund, and (c)–(g) vertical profiles of M_{BC} concentrations during the High-performance Instrumented Airborne Platform for Environmental Research (HIAPER) Pole-to-Pole Observations (HIPPO) campaigns 1–5. The vertical profiles of simulated M_{BC} are averages over 60° – 80° N, 140° – 170° W. The BASE simulation is a Group 1 simulation, and the $S = 0.1\%$ (global) and $S = 0.1\%$ (> 60° N) simulations are Group 2 simulations, as summarized in Table 1.

2016; Shen et al. 2017; Vignati et al. 2010). For example, model simulations using the BC-aging parameterization of Liu et al. (2011) showed good agreement with surface M_{BC} observations in the Arctic (Liu et al. 2011; Shen et al. 2017), and our sensitivity simulation using this parameterization (Param_Liu2011), a Group 2 simulation, shows good agreement with surface M_{BC} at two observation sites (Barrow and Ny-Ålesund; Fig. S1). In contrast, previous studies using higher S_{thre} values severely underestimated observed surface M_{BC} in the Arctic (Liu et al. 2016; Vignati et al. 2010), and this underestimation is consistent with our Group 1 simulations. The difference in surface M_{BC} between Group 1 and Group 2 is larger in winter and early spring than in summer (June–August): the mean M_{BC} difference between Group 1 and Group 2 is $14\text{--}28\text{ ng m}^{-3}$ (the Group 2/Group 1 M_{BC} ratio is 2.7–3.9) in winter and $2.7\text{--}3.1\text{ ng m}^{-3}$ (the Group 2/Group 1 M_{BC} ratio is 1.4–1.7) in summer at the two sites. The larger M_{BC} difference between Groups 1 and 2 in winter is likely because the contribution of BC transported a long distance from the midlatitudes is larger in wintertime Arctic, and BC remaining in the Arctic atmosphere without wet scavenged during the long-range transport is more sensitive in winter than in summer to how activation and wet removal processes in liquid and mixed-phase clouds are treated (Matsui and Moteki 2020).

This distinct difference in surface M_{BC} between Groups 1 and 2 is seen over the whole Arctic (defined as $>70^{\circ}$ N)

(Fig. 5a). Although Group 1 simulates lower surface M_{BC} with small seasonal variations, Group 2 simulates clear seasonal variations of M_{BC} with a maximum in winter. Similar to the results at Barrow and Ny-Ålesund, the difference in surface M_{BC} between Groups 1 and 2 is larger in winter than in summer in the whole Arctic. The mean M_{BC} difference between Group 1 and Group 2 is 25 ng m^{-3} (Group 2/Group 1 M_{BC} ratio is 2.4) in winter and 3.1 ng m^{-3} (the Group 2/Group 1 M_{BC} ratio is 1.5) in summer in the whole Arctic ($>70^{\circ}$ N). In this study, simulated latitudinal distributions of wintertime surface M_{BC} at low and middle latitudes ($<60^{\circ}$ N) are similar, within 30% except Param_Liu2011, in all model simulations (Fig. 5b). Surface M_{BC} in the Arctic, however, differs substantially among the nine model simulations (by a factor of 3.5 at $>70^{\circ}$ N and 4.9 at $>80^{\circ}$ N). This result indicates that the treatment of S_{thre} is a major cause of the discrepancy in lower tropospheric M_{BC} in the Arctic among model simulations.

The Base and $S = 0.1\%$ (> 60° N) simulations reproduce observed M_{BC} vertical profiles in the Arctic reasonably well in winter and spring (Figs. 4c–e). Comparisons of these two simulations with the $S = 0.1\%$ (global) case indicate that for simulations of near-surface M_{BC} in the Arctic the treatment of S_{thre} at high latitudes is more important than that at low and middle latitudes. This result shows that the representation of activation and wet removal processes in the Arctic is one of the

FIG. 5. Simulated surface M_{BC} concentrations: (a) monthly variations of surface M_{BC} concentrations in the Arctic ($>70^{\circ}\text{N}$) and (b) latitudinal dependence of surface M_{BC} concentrations in winter. Four simulations used a mixing-state-resolved representation (solid lines), and five simulations used various parameterizations (dotted lines) for BC-aging processes. Group 1 and Emis $\times 2(>60^{\circ}\text{N})$ simulation results are shown by squares. The ranges of Group 1 (the hydrophobic-to-hydrophilic conversion rate is faster with S_{thre} around 0.3%) and Group 2 (the hydrophobic-to-hydrophilic conversion rate is slower with S_{thre} around 0.1%) simulation results are shown by red and blue shading, respectively.

most important factors for accurate simulation of near-surface M_{BC} in the Arctic, which is consistent with the results of recent studies (e.g., Shen et al. 2017).

The hydrophilic M_{BC} fraction to total Arctic M_{BC} in winter is less than 50% (19%–48%) in Group 2 and greater than 50% (64%–94%) in Group 1 simulations (Fig. 6d). As a result, because smaller amounts of BC are activated and wet removed in Group 2 (compared with Group 1), the lifetime of Arctic BC, defined as the ratio of the BC burden in the Arctic to the BC deposition flux in the Arctic, is longer in Group 2 simulations (14–20 days) (Fig. 6c). The hydrophilic fraction and the lifetime of Arctic BC in Group 2 are generally consistent with previous studies showing a good agreement of surface M_{BC} with observations (Shen et al. 2017). The lifetime of Group 1 (6.0–10 days) is about half that of Group 2 simulations (Fig. 6c). These differences between Groups 1 and 2 are remarkable in winter, but M_{BC} concentrations and the lifetime of BC in summer are similar between the two groups (Fig. S2).

Because our model uses a detailed microphysics-based mixing-state-resolved approach for BC simulations, we can compare simulated BC particle sizes and mixing states with observations (obtained by SP2). The mass median diameter of the BC core is about 190 nm globally and 140 nm in the Arctic ($>70^{\circ}\text{N}$) in the BASE simulation (annual mean). These simulated values are generally consistent with surface and aircraft observations (150–200 nm in near-source regions and 150–300 nm in the Arctic; Moteki et al. 2012; Schulz et al. 2019; Schwarz et al. 2010; Sharma et al. 2017; Yoshida et al. 2020), although the simulated BC size is slightly smaller than the observed range in the Arctic. The mean number fraction of BC-containing particles to total (BC-containing + BC-free) particles is 15% globally and 30% in the Arctic in the BASE simulation, within the SP2 size range (annual mean). These simulated fractions are also generally consistent with observations (20%–30% in

East Asia, 10%–25% in Europe, and 5%–20% in the Arctic; Matsui et al. 2011a,b, 2013; Reddington et al. 2013; Sharma et al. 2017) although the simulated BC-containing fraction is higher than observations in the Arctic.

c. Theoretical relationship between S_{max} and S_{thre}

Constraining S_{max} in the Arctic is not easy because S_{max} depends strongly on seasonal variations of dynamical and physical processes, cloud type (e.g., liquid water or mixed-phase clouds), and aerosol concentrations. Because Arctic S_{max} is not well understood, we discuss two cases separately: Case 1 assumes that the true Arctic S_{max} for liquid clouds is low ($\sim 0.1\%$), and Case 2 assumes that it is high ($\sim 0.3\%$). These S_{max} values were chosen to be consistent with the range of S_{max} obtained in our BASE simulation (Fig. 2) and S_{thre} assumed in the model simulations (0.3% for Group 1 and 0.1% for Group 2). In the following, we examine the relationship between the S_{max} and S_{thre} . As described in the introduction, S_{thre} and S_{max} need to be the same to ensure the theoretically correct calculations of BC activation (Fig. 1).

1) CASE 1: LOW S_{MAX}

Group 2, using S_{thre} values of around 0.1%, reproduces surface M_{BC} in the Arctic reasonably well (Fig. 4). If Arctic S_{max} is actually around 0.1%, then S_{max} and S_{thre} in Group 2 are both around $\sim 0.1\%$ and consistent. Therefore, we can consider that the good agreement of Group 2 with the observed surface M_{BC} in the Arctic is because the $S_{\text{max}}-S_{\text{thre}}$ relationship is calculated correctly for BC activation (scenario i in Fig. 7). Because S_{thre} is about 0.3% and higher than the true S_{max} in Group 1, this corresponds to the case depicted in Fig. 1c, where BC-containing particles that should not be activated (S_{crit} between 0.1% and 0.3%) can form cloud droplets. As a result, the activated fraction and wet removal efficiency of BC

FIG. 6. Simulated BC statistics in the Arctic ($>70^{\circ}\text{N}$) during winter, averaged over the whole Arctic: (a) surface BC mass (M_{BC}) concentrations, (b) column (vertically integrated) M_{BC} concentrations, (c) BC lifetime, and (d) the hydrophilic M_{BC} fraction relative to the total M_{BC} concentration.

are overestimated, and M_{BC} is therefore underestimated, in terms of the $S_{\max}$ – S_{thre} relationship in Group 1 (scenario ii in Fig. 7). If the true $S_{\max}$ value in the Arctic is around 0.1%, then a low $S_{\max}$ value ($\sim 0.1\%$) should be calculated by global climate models and a low S_{thre} value ($\sim 0.1\%$) should be used for the Arctic in BC-aging parameterizations.

This story is simple, and previous BC modeling studies that obtained good agreement between simulated results and Arctic M_{BC} observations by assuming low S_{thre} ($\sim 0.1\%$) implicitly assumed that $S_{\max}$ is low ($\sim 0.1\%$) in the Arctic. However, a few recent observations have suggested that $S_{\max}$ values are higher than 0.1% in the Arctic (Koike et al. 2019; Leaitch et al. 2016). For example, aircraft observations over the Canadian Arctic obtained $S_{\max}$ values of 0.3%–0.6% in July (Leaitch et al. 2016), and a recent study at Ny-Ålesund estimated $S_{\max}$ to be 0.2%–0.8% in summer (Koike et al. 2019). The same study also found that, for clouds consisting of supercooled water droplets, cloud droplet number concentrations are similar to number concentrations of aerosols greater than 70 nm in diameter throughout the year (Koike et al. 2019). When we assume hygroscopic aerosol species (e.g., ammonium sulfate), the critical diameter of 70 nm for cloud activation corresponds to an S_{thre} value of $\sim 0.3\%$, and this fact suggests that $S_{\max}$ of around 0.3% is plausible for clouds consisting of water droplets at the observation site (30%–60% of clouds in winter and early spring at Ny-Ålesund; Koike et al. 2019). Mori et al. (2021) also found that S_{thre} is about 0.3% throughout the

year at Ny-Ålesund by comparing BC size distributions in the atmosphere and rainwater [using the method developed by Moteki et al. (2019)]. These observations and our model simulations (Fig. 2) suggest that Arctic $S_{\max}$ can be much higher than 0.1% throughout the year, and that the results shown in the next subsection (high $S_{\max}$ case) can be used not only in summer when most of the precipitation in the Arctic is produced in liquid-phase clouds but also in winter and spring when ice-containing clouds can be dominant in the Arctic.

2) CASE 2: HIGH $S_{\max}$

If the true Arctic $S_{\max}$ value is high ($\sim 0.3\%$), then $S_{\max}$ and S_{thre} ($\sim 0.1\%$) are not consistent in Group 2. This situation corresponds to that shown in Fig. 1b; thus, BC-containing particles that should form cloud droplets (S_{crit} between 0.1% and 0.3%) are not allowed to form cloud droplets. In this case, Group 2 underestimates BC activation and overestimates surface M_{BC} in the Arctic in terms of the $S_{\max}$ – S_{thre} relationship (scenario iii of Fig. 7). Although the BC activation calculation is theoretically incorrect, Group 2 reproduces the observed surface M_{BC} in the Arctic reasonably well. In contrast, even though Group 1 underestimates observed surface M_{BC} in the Arctic, it calculates the $S_{\max}$ – S_{thre} relationship correctly (scenario iv of Fig. 7). In this case, it is important to consider factors other than the activation and wet removal processes to improve simulations of M_{BC} in the Arctic.

FIG. 7. Responses of surface M_{BC} concentrations to $S_{\max}$ and S_{thre} in the Arctic. (left) The theoretical relationship between $S_{\max}$ and S_{thre} in the Arctic for four scenarios. The theoretical relationship between $S_{\max}$ and S_{thre} is correct when the actual $S_{\max}$ value in the atmosphere and the S_{thre} value assumed in the model are similar, i.e., in (i) and (iv). When S_{thre} is higher than $S_{\max}$, i.e., in (ii), M_{BC} should be lower (underestimated) than it would be if $S_{\max}$ and S_{thre} were similar. When S_{thre} is lower than $S_{\max}$, i.e., (iii), M_{BC} should be higher (overestimated) than it would be if $S_{\max}$ and S_{thre} were similar. (right) The agreement between observed and simulated surface M_{BC} concentrations in the Arctic. The Group 2 simulations show good agreement with observations, whereas the Group 1 simulations underestimate observed surface M_{BC} concentrations in the Arctic.

The lifetime and hydrophilic fraction of Arctic BC in the $\text{Emis} \times 2(>60^\circ\text{N})$ sensitivity simulation, which doubled anthropogenic BC emissions at high latitudes, are similar to those in the Base simulation (Fig. 6) because the treatment of activation and wet removal processes is the same in both simulations. However, surface M_{BC} and its seasonal variation in the $\text{Emis} \times 2(>60^\circ\text{N})$ simulation are similar to those in the Group 2 simulations (Fig. 5). In addition, the $\text{Emis} \times 2(>60^\circ\text{N})$ simulation reproduces observed surface M_{BC} in the Arctic better than the Base simulation (Fig. 4). Doubled anthropogenic BC emissions at high latitudes are possible when we consider the uncertainties in current BC emission inventories (Bond et al. 2013; Dong et al. 2019; Huang et al. 2015; Klimont et al. 2017; Stohl et al. 2013). Global anthropogenic BC emissions are almost the same (differing by 0.9% only) in the Base and $\text{Emis} \times 2(>60^\circ\text{N})$ simulations. These results show that Arctic M_{BC} in the lower troposphere is determined by a delicate balance among emissions, activation, and wet removal. If understanding of just one of these processes (e.g., $S_{\max}$ in the activation calculation) is insufficient, then the treatment of these processes by models is incorrect, even if the models reproduce observed surface M_{BC} concentrations in the Arctic.

If the latitudinal dependence of $S_{\max}$ in the Base simulation (Fig. 2) is realistic, then models using current BC parameterizations, which assume a single S_{thre} globally, cannot simulate Arctic M_{BC} with the correct $S_{\max}$ – S_{thre} relationship. When we use a low S_{thre} value globally (e.g., Param_0.1%), the model can calculate the $S_{\max}$ – S_{thre} relationship correctly at low and

middle latitudes, but it underestimates BC activation and wet removal and overestimates surface M_{BC} at high latitudes (in terms of the $S_{\max}$ – S_{thre} relationship). When a high S_{thre} value is used globally (e.g., Param_0.3%), the model can calculate the $S_{\max}$ – S_{thre} relationship correctly at high latitudes but overestimates BC activation and wet removal at low and middle latitudes. Because BC transport from the middle latitudes to the Arctic is underestimated in this treatment, M_{BC} in the Arctic is underestimated (in terms of the $S_{\max}$ – S_{thre} relationship). In fact, Param_0.1% (Param_0.3%) simulates higher (lower) surface M_{BC} than the Base simulation in the Arctic (Fig. 5). To consider the latitudinal dependence of $S_{\max}$ in BC activation properly, models should calculate spatial distributions of S_{thre} and its consistency with $S_{\max}$, as in our global model, or use a BC parameterization with a lower S_{thre} in low and middle latitudes and a higher S_{thre} in high latitudes. The Param_0.1%–0.3% simulation, which uses $S_{\text{thre}} = 0.1\%$ in low and middle latitudes ($<60^\circ\text{N}$) and $S_{\text{thre}} = 0.3\%$ in high latitudes ($>60^\circ\text{N}$), can simulate surface M_{BC} concentrations in the Arctic closer to the Base simulation results than either Param_0.1% or Param_0.3%; thus, $S_{\max}$ and S_{thre} can be represented consistently globally by a BC-aging parameterization that uses two S_{thre} values in parallel.

d. Implications for historical and future changes in Arctic BC

Global emissions of aerosols and their precursor species have been enhanced from preindustrial times to the present

FIG. 8. Responses of the lifetime of Arctic BC when emissions are reduced. (a) Two pathways of the impacts of emission reductions on the activation, wet removal, and lifetime of Arctic BC: the BC aging effect in the upper portion of (a) and the $S_{\max}$ effect in the lower portion of (a). (b),(c) The relationship between the lifetime of Arctic BC in winter and emission reductions for the Base (red) and $S = 0.3\%$ (>60°N) (black) simulations. Simulation results using preindustrial and future emissions (triangles and squares, respectively) are also shown. The $S = 0.3\%$ (>60°N) simulations show the response of the BC aging effect to emission reductions. The difference between the $S = 0.3\%$ (>60°N) and Base simulations corresponds to the response of the $S_{\max}$ effect to emission reductions.

day and their future changes strongly depend on the choice of future emission scenarios (Lund et al. 2019). Arctic aerosol concentrations in the past and future climate therefore can be considerably different from those in the present-day climate. For example, because BC emissions in preindustrial times were lower than present-day emissions, Arctic BC concentrations under preindustrial conditions may have been lower than present-day concentrations. Recent studies have shown that BC and sulfate have decreased since the mid-1990s in association with a reduction of emissions at high latitudes and an increase in precipitation rate (e.g., Sharma et al. 2019). This decreasing tendency may continue in the future because global BC emissions are projected to decrease in most future emission scenarios (Lund et al. 2019). If Arctic aerosol concentrations in the past and future climate are different from those in the present day, the following two effects play a key role in estimating the activation, wet removal, and lifetime of Arctic BC: 1) the BC aging effect and 2) the $S_{\max}$ effect (Fig. 8a). The BC aging effect increases (decreases) the lifetime of Arctic BC because the reduction (enhancement) of emissions and aerosol concentrations leads to slower (faster) aging, lower (higher) activation, and less (more) efficient wet removal of BC. In contrast,

the $S_{\max}$ effect has an opposite response because the reduction (enhancement) of aerosol concentrations increases (decreases) $S_{\max}$ and the resulting activation and wet removal amounts of BC. Many BC aging parameterizations can consider the BC aging effect (partly for condensation processes) but cannot treat the $S_{\max}$ effect.

To explore the sensitivity of the BC aging and $S_{\max}$ effects to Arctic BC reductions, we made additional sensitivity simulations, in which emissions of all aerosols and their precursor species except dust and sea salt were reduced to 10%, 20%, and 50% of present-day emissions globally, for the Base and $S = 0.3\%$ (>60°N) cases (S_{thre} is assumed to be <0.3% at >60°N). The Base simulations can consider both the BC aging and the $S_{\max}$ effects. The $S = 0.3\%$ (>60°N) simulations, which assume $S_{\max}$ (or S_{thre}) <0.3% at high latitudes (>60°N), can consider the BC aging effect only. The comparisons between the Base and $S = 0.3\%$ (>60°N) simulations enable us to understand the relative importance of the BC aging and $S_{\max}$ effects to Arctic BC. The reduction of the hydrophilic M_{BC} fraction in the Arctic with decreasing emissions reflects the slowdown of BC aging processes in both the Base and $S = 0.3\%$ (>60°N) simulations (Fig. 9a). The slowdown of BC

FIG. 9. Responses of the hydrophilic M_{BC} fraction and S_{max} to emission changes. (a) The relationship between the hydrophilic fraction of Arctic M_{BC} during winter and emission reductions in the Base (red) and $S = 0.3\%$ ($>60^{\circ}\text{N}$) (black) simulations. (b) Zonal-mean latitudinal distributions of S_{max} in the Base simulations using present-day emissions (red), 10% of present-day emissions (orange), preindustrial emissions (black), and future emissions (light blue).

aging is likely because the reduction of emissions leads to a slowdown of condensation aging caused by changes of the gas-particle partitioning of organic and nitrate aerosols as well as a slowdown of coagulation aging caused by a reduction of aerosol number concentrations. This result is consistent with our recent study showing that BC aging processes under the preindustrial condition are slower than those under the present-day condition (Matsui 2020). The enhancement of S_{max} especially at high latitudes is also seen clearly with decreasing emissions (Fig. 9b). These results show that both BC aging and S_{max} effects are important in our Arctic BC simulations.

In the $S = 0.3\%$ ($>60^{\circ}\text{N}$) simulations, which can consider the BC aging effect only, the lifetime of Arctic BC in winter increases by 29% and 56% when emissions are reduced by 80% and 90%, respectively (black lines in Fig. 8c). In the Base simulations, which consider both the BC aging and the S_{max} effects, the lifetime of Arctic BC in winter increases by 12% and 22% when emissions are reduced by 80% and 90%, respectively (red lines in Fig. 8c). These results show that the consideration of the BC aging effect only (as treated in many BC aging parameterizations) leads to too high sensitivity of the lifetime of Arctic BC to emission changes. The lifetime of Arctic BC is not strongly sensitive to emission changes when both the BC aging and S_{max} effects are treated properly. When we use emissions of aerosols and their precursors for the year 1750 (2100), the lifetime of Arctic BC in winter is enhanced by 46% (27%) through the BC aging effect, whereas it decreases by 34% (12%) through the S_{max} effect (Fig. 8).

4. Conclusions and discussion

The comparisons of the $S_{\text{max}}-S_{\text{thre}}$ relationship between Case 1 (low S_{max}) and Case 2 (high S_{max}) demonstrate that the consistency between S_{max} and S_{thre} , which has not been considered by most BC-modeling studies, is essential for reducing

uncertainties in BC simulations in the Arctic. Our results show that wintertime surface M_{BC} concentrations in the Arctic are reproduced well when S_{thre} is low ($\sim 0.1\%$, Group 2), and they are severely underestimated when S_{thre} is high ($\sim 0.3\%$, Group 1). If the actual S_{max} in the Arctic is around 0.1%, the BC activation in Group 2 is realistic because S_{max} and S_{thre} are both $\sim 0.1\%$ and consistent in the Arctic. Some recent observations and our model simulations, however, suggest that S_{max} in the Arctic can be much higher ($\sim 0.3\%$). In this case, Group 2 would obtain good agreement with observed surface M_{BC} in the Arctic, despite incorrect calculations of BC activation. These results indicate that without better understanding of S_{max} in the Arctic, we cannot judge whether good agreement with observed BC has been obtained by theoretically consistent calculations of BC activation. Our results also show that a spatially varying $S_{\text{max}}-S_{\text{thre}}$ relationship can be represented by using a particle-size and mixing-state-resolved representation or a BC-aging parameterization that uses two S_{thre} values in parallel.

Reducing the large uncertainty in observation-based S_{max} estimates in the Arctic is important for constraining the $S_{\text{max}}-S_{\text{thre}}$ relationship and BC budget in the Arctic. Because S_{max} depends on the updraft velocity and aerosol number concentrations in clouds, reducing their large uncertainties in models is also necessary. For example, a recent study has shown that the current uncertainties in updraft velocity in clouds produce fourfold to fivefold differences in the direct radiative effect of BC in the Arctic (Matsui and Moteki 2020). Second, aerosol number concentrations and their size distributions in the Arctic are mainly controlled by non-BC aerosols. Improved representation of microphysical processes related to their concentrations such as new particle formation, condensation, and coagulation, will also be a key to improve S_{max} estimation in the Arctic (Baccarini et al. 2020).

A better representation of S_{thre} and its consistency with S_{max} is necessary to obtain a more accurate understanding of

the balance among emissions, activation, and wet removal of BC in the Arctic and to reduce the large discrepancies of M_{BC} among current global models. The historical and future changes in S_{max} and their impacts on BC activation and wet removal processes should be calculated explicitly in Arctic BC simulations because both S_{max} and BC-aging speed depend strongly on historical and future changes in emissions. Our findings are an important step toward better simulations of BC and its impacts on atmospheric and snow/ice surface radiative effects, which may strengthen the snow-albedo feedback, in the past, present, and future Arctic climates.

Acknowledgments. This work was supported by the Ministry of Education, Culture, Sports, Science and Technology of Japan and the Japan Society for the Promotion of Science (MEXT/JSPS) KAKENHI Grant Numbers JP17H04709, JP16H01770, JP19H04253, JP19H05699, JP19KK0265, JP20H00196, and JP20H00638 and MEXT Arctic Challenge for Sustainability (ArCS, JPMXD1300000000) and ArCS-II (JPMXD1420318865) projects. This work was also supported by the Environment Research and Technology Development Fund 2-1703 (JPMEERF20172003) and 2-2003 (JPMEERF20202003) of the Environmental Restoration and Conservation Agency. We are grateful to Yutaka Kondo (National Institute of Polar Research in Japan) and Makoto Koike (University of Tokyo) for providing us the surface BC data at Barrow and Ny-Ålesund. The authors also thank the U.S. National Oceanic and Atmospheric Administration (NOAA) Black Carbon Group for providing us their BC data from aircraft measurements.

Data availability statement. Data used in this study are available upon request from the corresponding author (Hitoshi Matsui).

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