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Enhancement of snow albedo reduction and radiative forcing due to coated black carbon in snow

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Abstract. When black carbon (BC) is internally mixed with other atmospheric particles, BC light absorption is effectively enhanced. This study is the first to explicitly resolve the optical properties of coated BC in snow, based on core/shell Mie theory and a snow, ice, and aerosol radiative model (SNICAR). Our results indicate that a 'BC coating effect' enhances the reduction of snow albedo by a factor of 1.1–1.8 for a non-absorbing shell and 1.1–1.3 for an absorbing shell, depending on BC concentration, snow grain radius, and core/shell ratio. We develop parameterizations of the BC coating effect for application to climate models, which provides a convenient way to accurately estimate the climate impact of BC in snow. Finally, based on a comprehensive set of in situ measurements across the Northern Hemisphere, we find that the contribution of the BC coating effect to snow light absorption has exceeded that of dust over northern China. Notably, the high enhancements of snow albedo reductions by BC coating effect were found in the Arctic and Tibetan Plateau, suggesting a greater contribution of BC to the retreat of Arctic sea ice and Tibetan glaciers.

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

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1 Introduction

Snow is the most reflective natural substance at Earth's surface and covers more than 30% of global land area (Cohen and Rind, 1991). Snow albedo feedback is considered one of the major energy balance factors of the climate system. Previous observations have revealed that light-absorbing particles (LAPs; e.g., black carbon (BC), organic carbon (OC), and mineral dust) within snow can reduce snow albedo and enhance the absorption of solar radiation (Hadley and Kirchstetter, 2012). As a result, LAPs play a significant role in altering snow morphology and snowmelt processes, and therefore have important effects on local hydrological cycles and global climate (Qian et al., 2009).

Given the importance of the climate feedback caused by LAPs in snow, studies have developed snow radiative models and sought to improve our understanding of the influence of LAP-contaminated snow on climate. For example, Warren and Wiscombe (1980) developed a radiative forcing model based on Mie theory and δ-Eddington approximation, and reported that snow albedo in visible wavelengths could be reduced by 5%–15% when 1000 ng g^{-1} BC is present in snow. Flanner et al. (2007) developed a more comprehensive snow albedo model (the snow, ice, and aerosol radiation model; SNICAR) for multilayer snowpack using the two-stream radiative transfer solution. In addition to BC, the SNICAR model also accounts for the potential effects of dust particles and volcanic ash on snow albedo. Recent studies have indicated that snow grain shape also has an important influence on snow albedo (e.g., Dang et al., 2016).

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

Although efforts have been made to optimize snow albedo models, current models still suffer from major limitations. Studies have indicated that when BC in the atmosphere is coated with other aerosols it can significantly enhance light absorption via a lensing effect compared with uncoated BC, as investigated using model simulations (e.g., Jacobson 2001; Matsui et al., 2018) and experimental measurements (e.g., Cappa et al., 2012; Peng et al., 2016). Moreover, coated BC has been observed to exist for only a few hours after emission in some regions (Moteki et al., 2007; Moffet and Prather, 2009). Global aerosol models that simulate microphysical processes have indicated that most BC is mixed with other particles within 1–5 days (Jacobson, 2001) at all altitudes (Aquila et al., 2011). Hence, the climate impacts of BC must be evaluated in the context of the effect of coating on light absorption enhancement.

Although the BC coating effect on light absorption enhancement in the atmosphere is broadly acknowledged, little research has been carried out on snow albedo. Flanner et al. (2007) developed the first radiative transfer model to investigate the coating effect on snow albedo, using sulfate as the BC particle coating with a constant absorption enhancement factor of ~1.5. Subsequently, Wang at al. (2017) used a similar constant light absorption enhancement factor for their spectral albedo model for dirty snow (SAMDS). However, the factor varies with the optical properties of different coatings, the core/shell ratio, wavelength, and other parameters in real environments (Lack and Cappa, 2010; Liu et al., 2017). For example, Liu et al. (2017) reported that the core/shell ratio is a key control on light absorption enhancement. You et al. (2016) suggested that

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

light absorption enhancement is highly correlated with visible or near-infrared (NIR) wavelengths and coating material. Furthermore, a core/shell Mie theory simulation (Lack and Cappa, 2010) found light absorption enhancement was smaller for mildly absorbing coatings (e.g., OC) than non-absorbing coatings (e.g., sulfate). Hence, using a constant enhancement factor will result in biased simulation estimates, against refining our knowledge of the hydrological and climate impacts of BC in snow.

In this study we apply core/shell Mie theory to calculate the optical properties of BC coated with both mildly absorbing OC and non-absorbing sulfate, and use these results within a SNICAR model to evaluate the influence on snow albedo. Parameterizations for the BC coating effect are then developed for application in other snow albedo and climate models. Finally, we estimate the enhancements of snow albedo reductions and associated radiative forcing by the BC coating effect across the Northern Hemisphere, by combining model simulations with in situ observations of BC and OC concentrations in snow.

2 Methods

2.1 Modeling

2.1.1 Optical parameter calculations for coated BC

Figure 1a and 1c shows schematics of light absorption by externally and internally mixed particles (EMP and IMP, respectively). EMP refers to uncoated BC mixed with other particles, while IMP refers to BC that is assumed to be a core coated by another particle acting as a shell (Kahnert et al., 2012). For a non-absorbing shell, overall light

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

absorption includes contributions from the BC core and enhancement absorption from a lensing effect, while for an absorbing shell, the shell itself also contributes to light absorption.

To evaluate the BC coating effect on snow albedo, it is necessary to determine the optical parameters of coated BC. The refractive index (RI) of BC was assumed to be 1.95–0.79i following Lack and Cappa (2010), which is consistent with the original SNICAR model (Flanner et al., 2007). Two types of particle shells (non-absorbing and absorbing) were considered. The non-absorbing shell was represented using sulfate, and its RI was assumed to be 1.55–10^{-6}i following the atmospheric study of Bond et al. (2006). The absorbing shell was represented using OC, which is a major light-absorbing particle in snow (Wang et al., 2013). The RI of OC varies with wavelength. Here, a fixed mass absorption coefficient (MAC) for OC of 0.3 m^2 g^{-1} at 550 nm, a real RI of 1.55, and a particle diameter of 200 nm were assumed, following Lack and Cappa (2010). Based on Mie theory, an imaginary RI value of -1.36×10^{-2}i at 550 nm was calculated. Subsequently, wavelength-dependent imaginary RI values (Figure S1) were derived according to an absorption angstrom exponent (AAE) of −6 (Sun et al., 2007).

For a core/shell-structured particle, the core and shell diameters refer to the BC core diameter and the whole particle diameter, respectively. BC diameters are usually in the range of ~50–120 nm in the atmosphere (Corbin et al., 2018), and are typically larger by ~20 nm in snow due to a removal process via wet deposition (Schwarz et al., 2013). Therefore, we assumed the BC diameter in snow was 100 nm. Core and shell

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

diameters, RI, and wavelength were then used in a Mie model to derive optical parameters of the core/shell particle, including single scatter albedo (SSA), asymmetry factor (*g*), and extinction cross-section (Q_{ext}). The mass extinction coefficient (MEC) of the core/shell particle was calculated based on Q_{ext} and the density, given as 1.8 g cm^{-3} for BC (Bond et al., 2006), 1.2 g cm^{-3} for sulfate, and 1.2 g cm^{-3} for OC (Turpin and Lim, 2001).

2.1.2 Snow albedo calculations

We simulated snow albedo with the SNICAR model (Flanner et al., 2007), which calculates the radiative transfer in snowpack based on the theory of Warren and Wiscombe (1980) and a two-stream multilayer radiative approximation (Toon et al., 1989). Here, we summarize only the model features in SNICAR that are crucial to our study. SNICAR allows for a vertical multilayer distribution of snow properties, LAPs, and heating throughout the snowpack column. Input optical parameters (MEC, SSA, and *g*) of snow grains and BC were calculated off-line using Mie theory. SNICAR provides albedo changes from uncoated and sulfate-coated BC on snow, as well as dust particles and volcanic ash (for further details, see Flanner et al., 2007).

For SNICAR snow albedo simulations of uncoated BC, concentrations of both BC and other particles were input directly. For coated BC, optical parameters (MEC, SSA, and *g*) of IMP (calculated above) were first archived as lookup tables within SNICAR, and then the concentration of IMP was input.

2.2 Calculation of broadband snow albedo

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

The spectral albedo (α_λ) was integrated over the solar spectrum (λ = 300–2500 nm) and weighted by the incoming solar irradiance (S_λ) to calculate broadband snow albedo ($\alpha_{integrated}$):

$$\alpha_{\text{integrated}} = \frac{\int \alpha_\lambda S_\lambda d_\lambda}{\int S_\lambda d_\lambda} \quad (1)$$

The incoming solar irradiance was a typical surface solar spectrum for mid–high latitudes from January to May, calculated with the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Pu et al., 2019), which is one of the most widely used models for radiative transfer simulations (for further details, see Ricchiazzi et al. 1998).

2.3 Parameterizations

In the original SNICAR model, the BC coating effect is simply parameterized with an absorption enhancement of ~1.5 (Flanner et al., 2007). However, the effect of BC coating on snow albedo is widely variable and dependent on BC concentration, core/shell ratio, snow grain size, and the type of particle shell (see Section 3.3). In view of this complexity, more explicit parameterizations were developed in this study:

$$\mathrm{E}_{\alpha,\text{integrated}} = \frac{\alpha_{int,\text{integrated}}}{\alpha_{ext,\text{integrated}}} \quad (2)$$

where $\alpha_{ext,\text{integrated}}$ and $\alpha_{int,\text{integrated}}$ are broadband snow albedos for EMP and IMP, respectively. Following a previous empirical formulation (Hadley and Kirchstetter, 2012), $\mathrm{E}_{\alpha,\text{integrated}}$ was parameterized as

$$\mathrm{E}_{\alpha,\text{integrated,para}} = a_0 \times (C_{BC})^{a_1} + a_2 \quad (3)$$

$$a_1 = b_0 \times (\log_{10}(R_{ef}/50))^{b_1} \quad (4)$$

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

where $E_{\alpha,\mathrm{integrated,para}}$ is the parameterized $E_{\alpha,\mathrm{integrated}}$, C_{BC} is the BC concentration, and R_{ef} represents the snow grain radius. The terms a_0, a_1, a_2, b_0, and b_1 are the empirical coefficients and dependent on the core/shell ratio and the type of particle shell. To enhance the precision, parameterizations were divided into two groups: the first to account for relatively clean snow (with BC concentrations < 200 ng g^{-1}) and the second for relatively polluted snow (200 ng g^{-1} < BC concentrations < 1000 ng g^{-1}).

2.4 Calculation of in situ snow albedo and radiative forcing

In situ broadband clear-sky ($\alpha_{\mathrm{integrated}}^{\mathrm{clear,in-situ}}$) and cloudy-sky ($\alpha_{\mathrm{integrated}}^{\mathrm{cloudy,in-situ}}$) albedos were calculated separately based on in situ snow-LAP parameters and SBDART simulated clear-sky and cloudy-sky incoming solar irradiance by assuming a semi-infinite snowpack. The in situ all-sky albedo ($\alpha_{\mathrm{integrated}}^{\mathrm{all-sky,in-situ}}$) was then calculated using weighted clear-sky and cloudy-sky albedos depending on cloud fraction (CF), given as:

$$\alpha_{\mathrm{integrated}}^{\mathrm{all-sky,in-situ}} = \mathrm{CF} \times \alpha_{\mathrm{integrated}}^{\mathrm{cloudy,in-situ}} + (1-\mathrm{CF}) \times \alpha_{\mathrm{integrated}}^{\mathrm{clear,in-situ}} \quad (5)$$

In situ radiative forcing by LAPs was calculated by multiplying the derived broadband albedo reduction by the downward shortwave flux at the snow surface (Dang et al., 2017). Figure S2 shows the spatial distributions of solar flux and cloud fraction, which were obtained from the Clouds and the Earth's Radiant Energy System (CERES) (https://ceres.larc.nasa.gov/products.php?product=SYN1deg).

3 Results and discussion

3.1 Impact on particle light absorption

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

Figure 1b and 1d shows light absorption ratios (light absorption enhancement, E_{abs}) for IMP versus EMP. Based on Bond et al. (2006), we show the most common core/shell ratios (the ratio of the diameter of the whole particle to the BC core) of 1.2, 1.5, 2.0, and 2.5 in real environments to represent the thickness of shells, and we used detailed core/shell ratios from 1.1 to 3.0 (in intervals of 0.1) for parameterizations (see Section 3.5). E_{abs} varies with the wavelength and increases with core/shell ratio, in contrast to the default E_{abs} value used in the original SNICAR model, which remains constant. For a non-absorbing shell, the light absorption of IMP is larger than EMP across all wavelengths (300–1400 nm). For an absorbing shell, E_{abs} is similar to the non-absorbing shell in NIR, but becomes smaller in visible (VIS) light and ultra-violet (UV), which implies the absorbing shell reduces whole-particle light absorption and contributes negatively to E_{abs}. A plausible explanation is that the shell absorbs incident photons, causing fewer to reach the core. In such a case, the additional photons absorbed by the shell are fewer than the reduced number of photons absorbed by the lensing effect and the BC core. Furthermore, the absorbing shell reduces E_{abs} to <1 in UV at high core/shell ratios, implying that the lensing effect absorption at those wavelengths cannot recover the BC core absorption reduction, resulting in fewer photons reaching the core.

3.2 Impact on spectral snow single-scattering properties and albedos

In real snowpack, BC can effectively enhance snow single-scattering co-albedo (1–ω), but its effect on other snow optical parameters, such as the asymmetry factor

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

and extinction efficiency, is negligible (He et al., 2017). Therefore, we focus our discussion on coating-induced enhancement of snow single-scattering co-albedo ($E_{1-\omega}$), snow albedo (E_{α}), and snow albedo reduction ($E_{\Delta\alpha}$).

The coating effect has a clear influence on $E_{1-\omega}$ and E_{α} in UV and VIS, but affects $E_{\Delta\alpha}$ across all wavelengths (Figure 2). The core/shell ratio makes a positive contribution to $E_{1-\omega}$ and $E_{\Delta\alpha}$, and a negative contribution to E_{α}. The BC concentration makes a positive contribution to $E_{1-\omega}$ and a negative contribution to $E_{\Delta\alpha}$ and E_{α}. For a given BC concentration and core/shell ratio, $E_{1-\omega}$ and $E_{\Delta\alpha}$ decrease, and E_{α} increases, with longer wavelength from UV to VIS. In addition, the absorbing shell shows a negative influence for $E_{1-\omega}$ and $E_{\Delta\alpha}$, but positive for E_{α} compared with a non-absorbing shell, particularly under UV. Moreover, for an absorbing shell, $E_{1-\omega}$ and $E_{\Delta\alpha}$ are <1, and E_{α} is >1 at approximately <350 nm in the case of high core/shell ratios because of lower light absorption by IMP than EMP at those wavelengths.

3.3 Impact on broadband snow single-scattering properties and albedos

Compared with spectral optical properties, our broadband results have wider implications for the research community. Core/shell ratios dominate variations in spectrally weighted $E_{1-\omega,\ \text{integrated}}$, $E_{\alpha,\ \text{integrated}}$, and $E_{\Delta\alpha,\ \text{integrated}}$ (Figure 3). Snow grain size and BC concentration show a negative contribution to $E_{\alpha,\ \text{integrated}}$, but barely affect $E_{\Delta\alpha,\ \text{integrated}}$. However, the type of particle shell has a significant impact on $E_{1-\omega,\ \text{integrated}}$, $E_{\alpha,\ \text{integrated}}$, and $E_{\Delta\alpha,\ \text{integrated}}$. Compared with $E_{1-\omega,\ \text{integrated}}$ and $E_{\alpha,\ \text{integrated}}$, $E_{\Delta\alpha,\ \text{integrated}}$ has a more direct influence on radiative forcing and climate change. $E_{\Delta\alpha,\ \text{integrated}}$ largely falls

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

within a range of 1.11 to ~1.80 for a non-absorbing shell, which exceeds the values for an absorbing shell (1.10 to ~1.33) by a factor of 1.0–1.35 (with BC concentrations within 1000 ng g^{-1}, a snow grain radius of 100–500 µm, and core/shell ratios from 1.2 to 2.5). In contrast, the $E_{\Delta\alpha,\ \mathrm{integrated}}$ from the original SNICAR model only shows a small variation of 1.23–1.31. This is similar to a non-absorbing shell with a core/shell ratio of ~1.5, which implies the original SNICAR model only reflects a coating effect on snow albedo reduction at an intermediate core/shell ratio.

3.4 Uncertainties

Although the imaginary RI value of OC has been theoretically calculated (Section 2.1), we note that in real snowpack there is large uncertainty because the types and optical properties of OC varies spatially and temporally due to different emission sources and photochemical reactions in the atmosphere (e.g. Lack and Cappa, 2010). To address this issue, we tested the degree of influence of imaginary RI on $E_{\alpha,\ \mathrm{integrated}}$, and $E_{\Delta\alpha,\ \mathrm{integrated}}$ values by increasing and decreasing the calculated imaginary RI by 50% (Figure S1), which studies have shown to be plausible (e.g., Lack et al., 2012). We find imaginary RI uncertainty to be ±1% for $E_{\alpha,\ \mathrm{integrated}}$ and ±5% for $E_{\Delta\alpha,\ \mathrm{integrated}}$.

In addition, observations show large variation in the size distribution of atmospheric and snow BC particles (Schwarz et al., 2013), which can affect snow optical properties and albedo (He at al., 2018). Therefore, we examined the effects of BC particle size on $E_{\alpha,\ \mathrm{integrated}}$ and $E_{\Delta\alpha,\ \mathrm{integrated}}$ with two additional BC diameters of 50 nm and 150 nm, which are within observed size ranges (Schwarz et al., 2013) and are

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

The Cryosphere Discussions
Open Access
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comparable to BC particle sizes used in other studies (e.g. He et al., 2018). We find the uncertainty attributed to BC diameter is ±1% for $E_{\alpha,\ \mathrm{integrated}}$ and ±13% for $E_{\Delta\alpha,\ \mathrm{integrated}}$. Overall, the total uncertainty related to imaginary RI and BC diameter is ±1.4% for $E_{\alpha,\ \mathrm{integrated}}$ and ±13.9% for $E_{\Delta\alpha,\ \mathrm{integrated}}$.

3.5 Parameterizations of the coating effect

Figure 4 compares parameterized $E_{\alpha,\ \mathrm{integrated,\ para}}$ with SNICAR-modeled $E_{\alpha,\ \mathrm{integrated}}$, and Tables S1 and S2 list the empirical coefficients (see Section 2.3) derived from nonlinear regression processes. $E_{\alpha,\ \mathrm{integrated,\ para}}$ and $E_{\alpha,\ \mathrm{integrated}}$ show a strong correlation, with R^2 = 0.988 (0.986) for a non-absorbing shell and R^2 = 0.987 (0.986) for an absorbing shell in relatively clean (polluted) snow, and root mean squared errors of 1.81×10^{-3} (4.70×10^{-3}) and 1.41×10^{-3} (3.76×10^{-3}), respectively. Biases for $E_{\alpha,\ \mathrm{integrated,\ para}}$ are smallest for intermediate BC concentrations and snow grain radius, but become larger at extremely low or high values, due mainly to processes within the nonlinear regression method. Overall, the $E_{\alpha,\ \mathrm{integrated}}$ can be well reproduced by $E_{\alpha,\ \mathrm{integrated,\ para}}$ for both a non-absorbing and absorbing shell under the conditions of both relatively clean and polluted snow.

Therefore, other studies can estimate the coating effect of BC on snow albedos and radiative forcing very conveniently by combining the original SNICAR or other snow radiative forcing model with our new parameterizations, which can reduce snow albedo simulation bias. For example, Wang et al. (2017) compared observations and model simulations of snow albedo reduction in northern China. They found simulated

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

results overestimated snow albedo by ~0.06 in polluted snow, despite considering the effects of all LAP types (BC, OC, and dust), snow grain type, and snow depth. However, if the coating effect is taken into account and the new parameterizations are applied (according to their measured LAPs and snow parameters), the difference between simulation and observation is reduced to ~0.02, thereby improving the simulation. Furthermore, although most global climate models (GCMs) account for internal mixing of atmospheric BC, they barely consider the coating effect for BC in snow (Bond et al., 2013). In addition, different GCMs apply different types of snow radiative transfer models, which means that one physical mechanism responsible for the BC coating effect in snow cannot be suitable for all GCMs. In this case our parameterizations are particularly helpful, as they are easily used in any GCM and improve understanding of how BC in snow influences local hydrological cycles and global climate.

3.6 Measurement-based estimate of coating effect

To evaluate the coating effect of BC on both snow albedo and radiative forcing in real snowpack, we collected in situ measurements of BC and OC concentrations in snow (Figure 5) during field campaigns in the Arctic in spring 2007–2009 (Doherty et al., 2010), North America in January–March 2013 (Doherty et al., 2014), northern China in January–February 2010 and 2012 (Ye et al., 2012; Wang et al., 2013), and the Tibetan Plateau in spring 2010 and 2012 (Wang et al., 2013; Li et al., 2017, 2018; Pu et al., 2017; Zhang et al., 2017, 2018). Measurements are separated into four geographical regions (Figure 5c): the Arctic, North America (NA), northern China (NC),

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

and the Tibetan Plateau (TP). An absorbing shell of OC was assumed in measured snowpack, which is plausible because previous studies have found that OC is the dominant coating in the atmosphere (e.g., Cappa et al., 2012). The OC/BC mass ratio is generally from 1 to 10, with the corresponding core/shell ratio from 1.3 to 2.5 (Figure 5b). The average core/shell ratio was highest (2.45) in the TP, followed by 1.92 and 1.81 in the Arctic and NC, respectively, and lowest (1.31) in NA (Figure 5d). These results reveal the BC coating effect had a larger impact on snow albedo in the TP than in other regions.

Figure 6 presents the coating effect on snow albedo reduction and radiative forcing based on measured BC in real snowpack, and Figures S3–S5 show spatial distributions and statistics. For fresh snow, we find that IMP results in greater snow albedo reductions compared with EMP by factors of 1.27, 1.19, 1.13, and 1.23 in the TP, NC, NA, and the Arctic, respectively (Figure 6a and b). Correspondingly, we find that IMP leads radiative forcing by 1.27, 1.20, 1.14, and 1.22 for these same regions. The highest (lowest) enhancement was found in the TP (NA), which corresponds to the highest (lowest) OC/BC mass ratio and core/shell ratio in the TP (NA). For old snow, the enhancement of snow albedo reductions due to the BC coating effect are 1.24, 1.15, 1.13, and 1.23 in TP, NC, NA, and the Arctic, respectively. The corresponding radiative forcing reductions are 1.24, 1.16, 1.14, and 1.22. The enhancement shows a slight decrease with snowpack aging, which is consistent with the results in Figure 3. Of note, we found the contribution of coating effect to light absorption has exceeded dust over

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

most areas of northern China after comparing with previous studies of dust in snow (Wang et al., 2013, 2017; Pu et al., 2017), which further demonstrated the critical role of BC coatings in snow albedo evaluation.

In contrast to previous studies, we note that enhanced light absorption in snow from the BC coating effect should be taken into account, especially in the Arctic and the TP. Arctic sea ice has shown a sharp decline in recent decades (Ding et al., 2019), and climate models predict a continued decreasing trend (Liu et al., 2020) that is likely to perturb the Earth system and influence human activities (Meier et al., 2014). Multi-model ensemble simulations indicate that greenhouse gases cannot fully explain this decline, and recent studies have proposed that BC deposition in snow and sea ice is an important additional contributor (e.g., Ramanathan and Carmichael, 2008). Furthermore, the TP holds the largest ice mass outside polar regions, and acts as a water 'storage tower' for more than 1 billion people in South and East Asia. Tibetan glaciers have rapidly retreated over the last 30 years (Yao et al., 2012), raising the possibility that many glaciers and their fresh water supplies could disappear by the middle of the 21st century. Observed evidence suggests that BC deposition is a significant contributing factor to this retreat (Xu et al., 2009), but quantitatively modeling the effect of BC on glacier dynamics is a challenge, partly because of incomplete radiative transfer mechanisms within models. Due to the significant contribution of BC to retreating Arctic sea ice and Tibetan glaciers, and the strong enhancement of light absorption by coated BC, the coating effect must now be considered in climate models

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

that are designed to accurately reconstruct both the historical record and future change.

4 Conclusions

This study evaluated the effect of BC coating on snow albedo and radiative forcing by combing core/shell Mie theory and the snow-albedo model SNICAR. We found that the coating effect reduces snow albedo by a factor of 1.11–1.80 for a non-absorbing shell and 1.10–1.33 for an absorbing shell, when BC concentrations are within 1000 ng g^{-1}, the snow grain radius is 100–500 μm, and the core/shell ratio is 1.2–2.5. The core/shell ratio plays a dominant role in reducing snow albedo. Furthermore, an absorbing shell causes a smaller snow albedo reduction than a non-absorbing shell because of a lensing effect, whereby the absorbing shell reduces photon absorbance in the BC core. Our results can effectively account for the complex enhancement of snow albedo reduction due to coating effect in real environments.

Parameterizations for the coating effect are further developed for use in snow albedo and climate models. Parameterized and simulated results show strong correlations for both clean and polluted snowpack. The root mean squared error of parameterized $E_{\alpha,\ \mathrm{integrated,\ para}}$ is low (1.41×10^{-3}). A list of empirical coefficients for parameterizations is provided for most seasonal snowpack field cases, with BC concentrations within 1000 ng g^{-1}, snow grain sizes from 100–1000 μm, and core/shell ratios from 1.1 to 3.0. We demonstrate that parameterizations can reduce simulation bias for local experiments in snow albedo models and, more importantly, can be applied to GCMs to improve our understanding of how BC in snow affects local hydrological

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

cycles and global climate.

Based on a comprehensive set of field measurements across the Northern Hemisphere, the BC coating effect in real snowpack was evaluated by assuming the presence of an absorbing OC shell. The enhancement of snow albedo reduction was 1.133–1.273 and enhancement of radiative forcing was 1.135–1.274, which exceeds the contribution of dust to snow light absorption over most areas of northern China. Of note, the greatest enhancements were detected on the Tibetan Plateau and in the Arctic, which will likely contribute to further Arctic sea ice and Tibetan glacier retreat. Our findings indicate that the coating effect must be considered in future climate models, in particular to more accurately evaluate the climate of the Tibetan Plateau and the Arctic.

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

Conflict of interest

The authors declare that they have no conflict of interest.

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Author contributions

X Wang and W Pu invited the project. W Pu and X Wang designed the study. W Pu wrote the paper with contributions from all co-authors. TL Shi processed and analyzed the data.

https://doi.org/10.5194/tc-2020-238
Preprint. Discussion started: 2 September 2020

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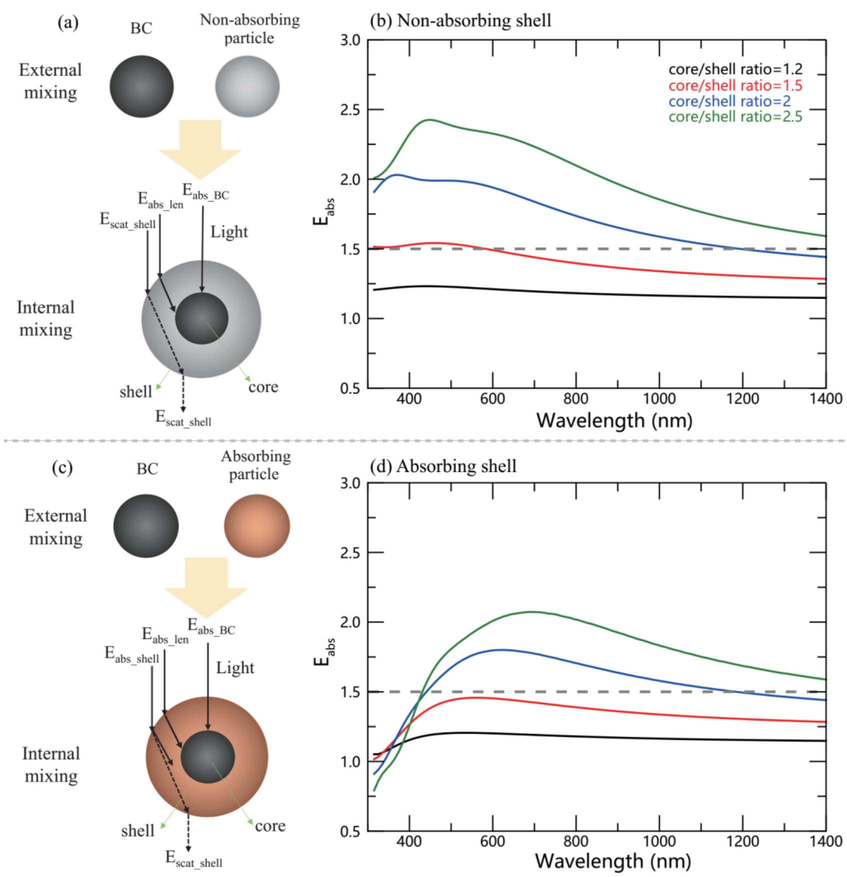

Figure 1. Schematic diagrams showing the light absorption for an external mixture and internal mixture of BC, for (a) a non-absorbing particle and (c) an absorbing particle. Also shown is the enhancement of light absorption from the internal mixture (E_{abs}) compared to the external mixture of BC with (b) non-absorbing and (d) absorbing particles. The internal mixed particle was assumed to be a core/shell structure with a black carbon (BC) core.

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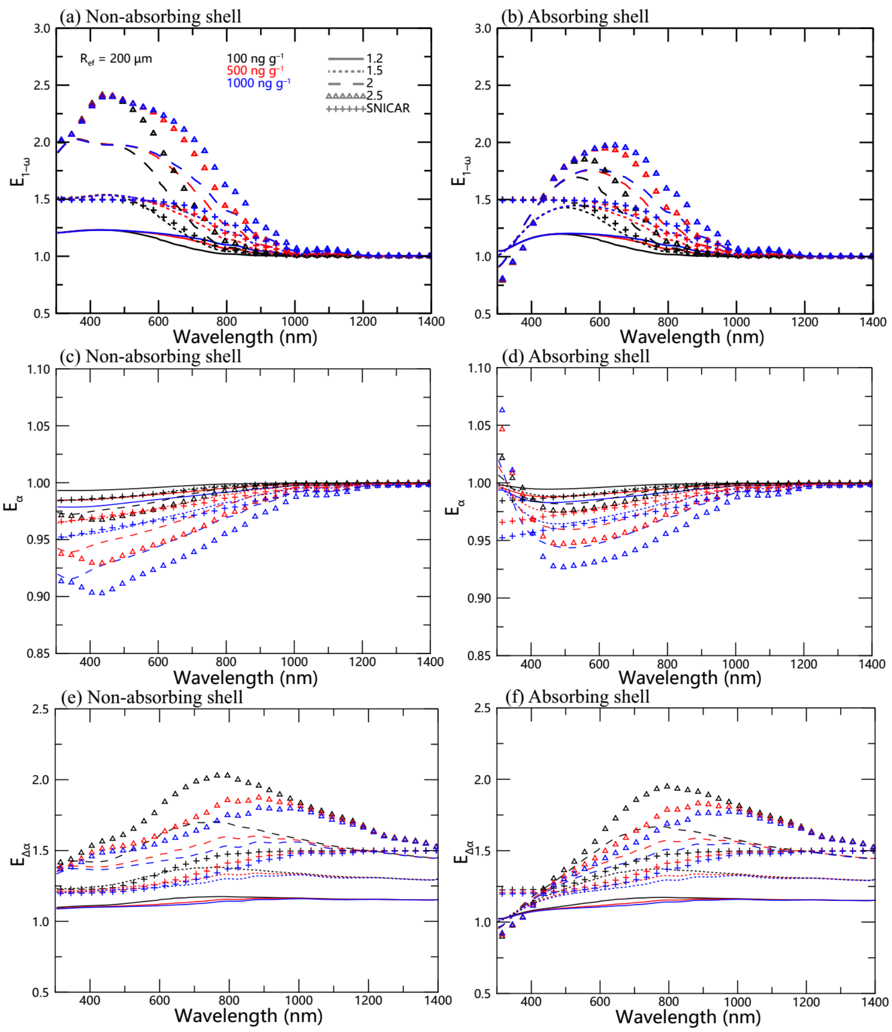

Figure 2. Ratios of snow single-scattering co-albedo ($E_{1-\omega}$) from an internal mixed particle to an external mixed particle as a function of wavelength, with different BC concentrations and core/shell ratios for (a) a non-absorbing shell, and (b) an absorbing shell. (c) and (d) Same as (a) and (b), but for snow albedo (E_{α}). (e) and (f) Same as (a) and (b), but for snow albedo reduction ($E_{\Delta\alpha}$). The snow grain radius was assumed to be 200 nm.

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Preprint. Discussion started: 2 September 2020

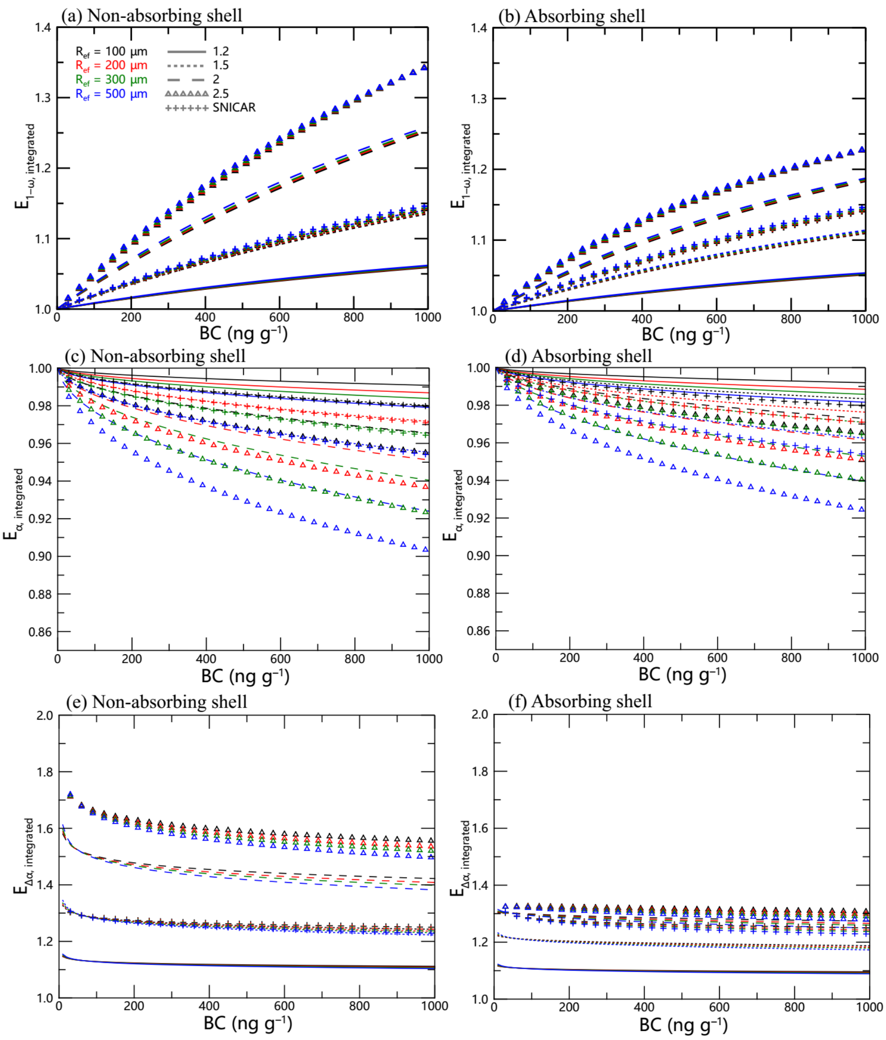

Figure 3. The spectrally weighted $E_{1\text{-}\omega}$ ($E_{1\text{-}\omega,\ \text{integrated}}$) over 300–1400 nm of a typical surface solar spectrum at mid–high latitude from January to May, for (a) a non-absorbing shell and (b) an absorbing shell as a function of black carbon (BC) concentration with different snow grain radii and core/shell ratios. (c) and (d) Same as (a) and (b), but for broadband snow albedo ($E_{\alpha,\ \text{integrated}}$). (e) and (f) Same as (a) and (b), but for broadband snow albedo reduction ($E_{\Delta\alpha,\ \text{integrated}}$).

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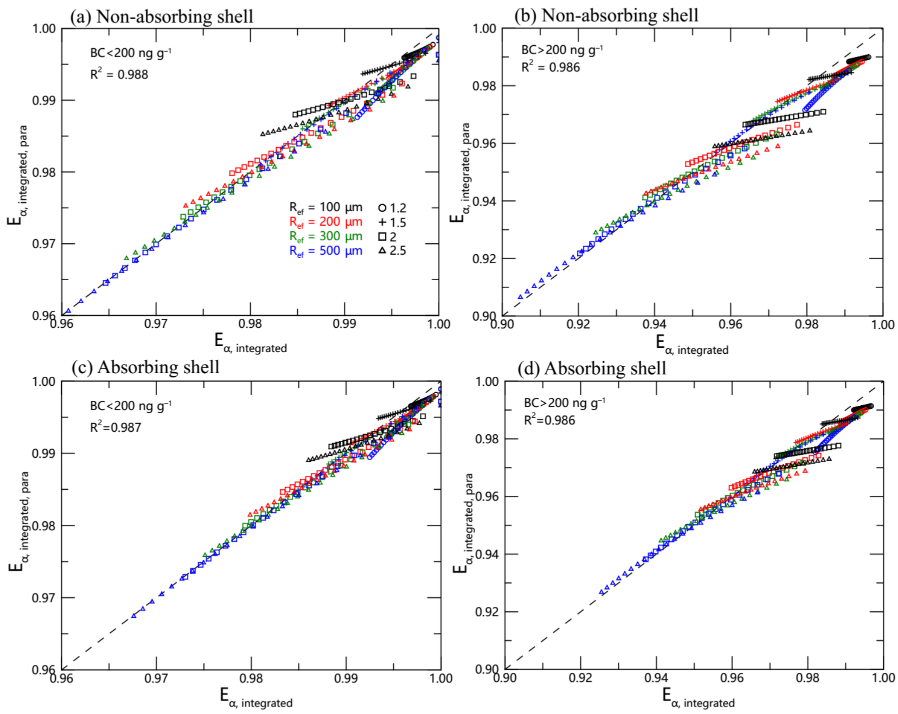

Figure 4. Comparisons of model calculated $E_{\alpha,\ \mathrm{integrated}}$ and parameterized $E_{\alpha,\ \mathrm{integrated,\ para}}$ for (a) relatively clean snow (BC concentration <1000 ng g^{-1}), and (b) relatively polluted snow (BC concentration >1000 ng g^{-1}) for a non-absorbing shell. (c) and (d) Same as (a) and (b), but for an absorbing shell.

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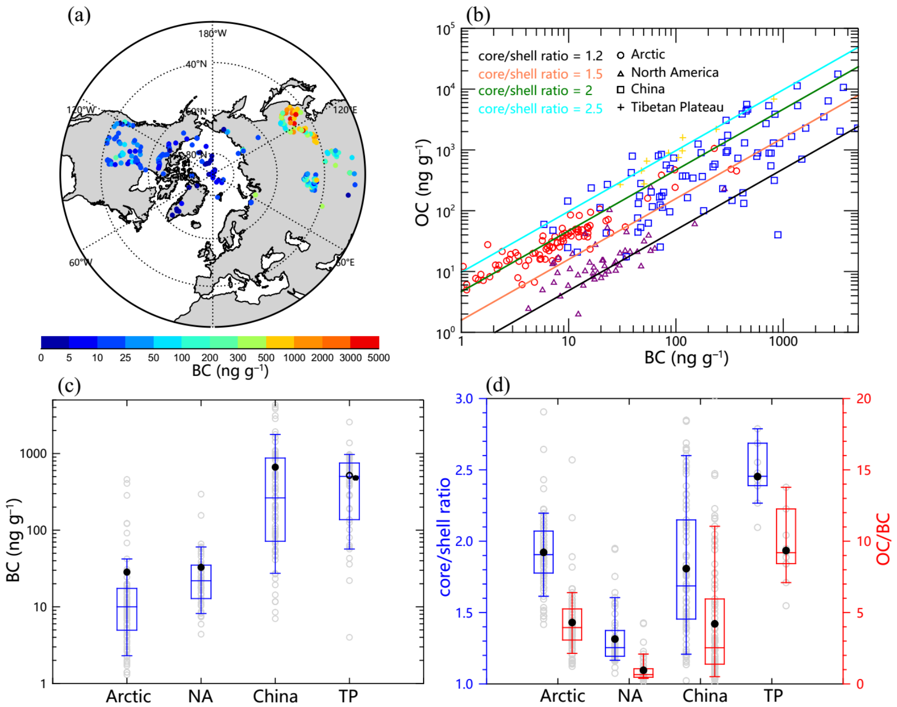

Figure 5. (a) The spatial distribution of measured black carbon (BC) concentrations across the Northern Hemisphere. (b) Comparison of BC and organic carbon (OC) concentrations in: the Arctic, North America (NA), northern China (NC) and the Tibetan Plateau (TP). (c) Statistical plots of BC concentrations in different regions. The boxes denote the 25th and 75th quantiles, the horizontal lines denote the 50th quantiles (medians), solid dots denote averages, and whiskers denote the 10th and 90th quantiles. In situ data is shown as gray circles. (d) Same as (c) but for a core/shell ratio and OC/BC mass ratio, assuming a core/shell structure with a BC core and an absorbing OC shell.

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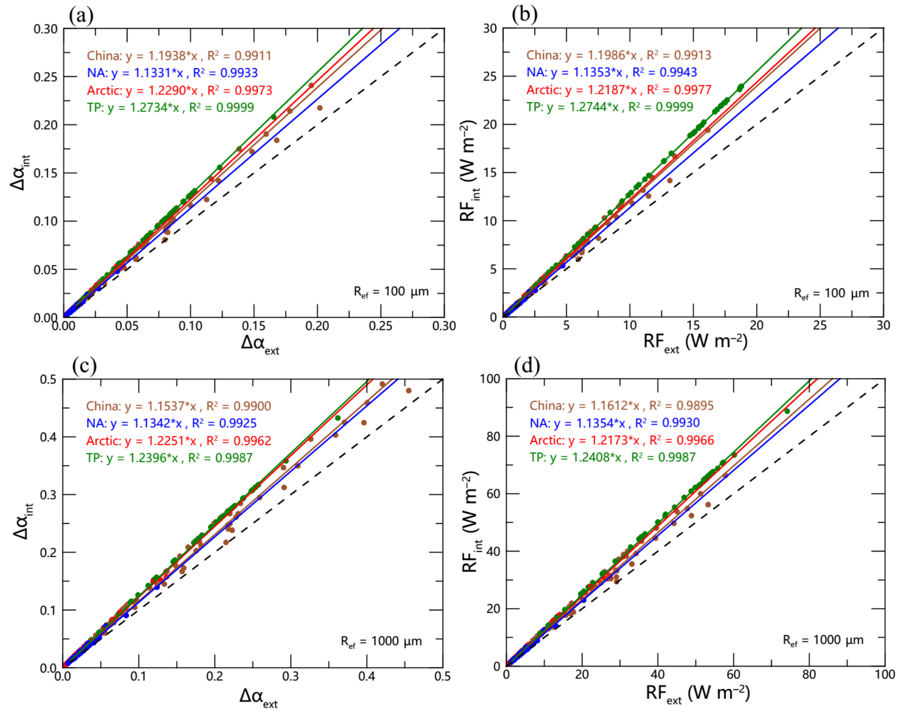

Figure 6. Comparisons of (a) the snow albedo reduction and (b) the radiative forcing by an internal mixed particle versus an external mixed particle, based on in situ measurements of fresh snow (assuming a snow grain radius of 100 μm). (c) and (d) Same as (a) and (b), but for old snow and assuming a snow grain radius of 1000 μm.