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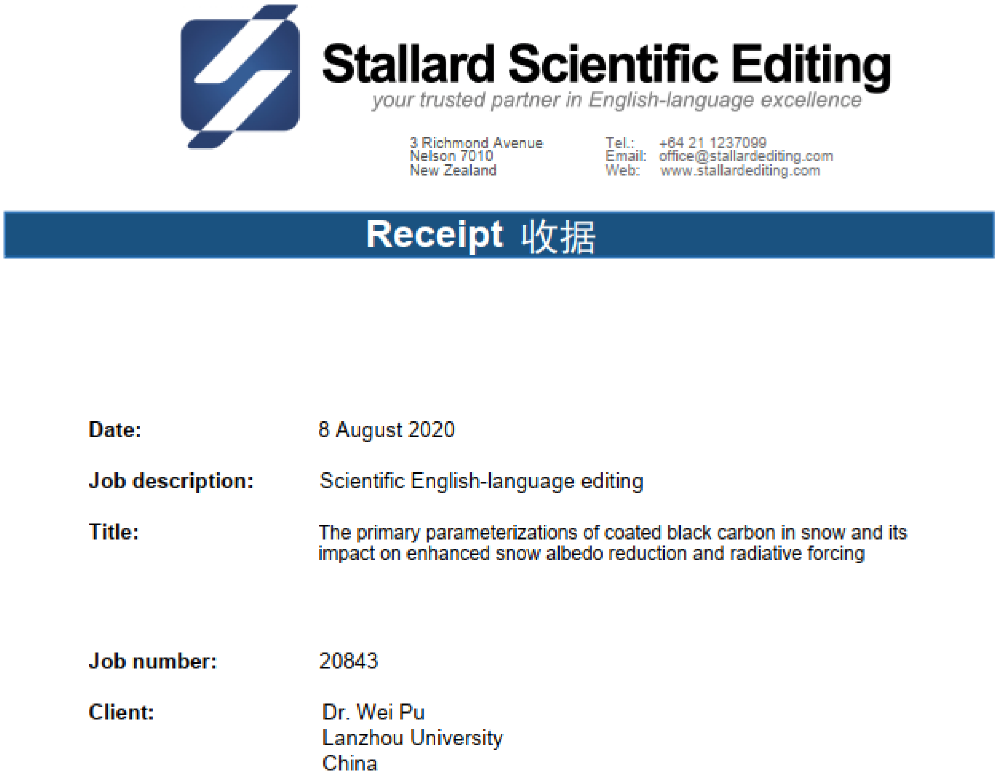

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# Enhancement of snow albedo reduction and radiative forcing due to coated black carbon in snow

Wei Pu[1], Tenglong Shi[1], Jiecan Cui[1], Yang Chen[1], Yue Zhou[1], Xin Wang[1,2]

[1]Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, College of Atmospheric Sciences, Lanzhou University, Lanzhou 730000, China

[2]Institute of Surface-Earth System Science, Tianjin University, Tianjin 300072, China

Corresponding author: Xin Wang (wxin@lzu.edu.cn)

**Abstract**~~.~~ When black carbon (BC) is ~~internally~~ mixed internally with other atmospheric particles, the BC light absorption effect is ~~effectively~~ enhanced. This study explicitly resolved the optical properties of ~~snow~~ coated ~~in~~ BC in snow ~~in snow,~~ based on the core/shell Mie theory and ~~a~~ the snow, ice, and aerosol radiative model (SNICAR). Our results indicated that ~~a~~ the ~~‘BC~~BC coating ~~effect’~~effect enhances the reduction ~~of~~in snow albedo by a factor ~~of~~ranging from 1.1–1.8 for a ~~non-absorbing~~nonabsorbing shell and 1.1–1.3 for an absorbing shell, depending on the BC concentration, snow grain radius, and core/shell ratio. We developed 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~~ areas~~. Finally, based on a comprehensive set of in situ measurements across the Northern Hemisphere, we ~~find~~ determined that the contribution of the BC coating effect to snow light absorption ~~has exceeded~~exceeds that of dust over northern China. Notably,~~ the~~ high enhancements ~~of~~~~in~~of snow albedo reduction~~s~~ ~~by~~ due to the 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.

## 1 Introduction

Snow is the most reflective natural substance ~~at~~ on the surface of Earth~~'s~~ ~~surface~~ and covers more than 30% of the 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~~ may reduce snow albedo and enhance the absorption of solar radiation (Hadley and Kirchstetter, 2012). As a result, LAPs play a ~~significant~~ major role in ~~altering~~ the alteration of snow morphology and snowmelt processes~~,~~ and therefore ~~have~~ yield 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 the Mie theory and the δ-Eddington approximation~~,~~ and reported that snow albedo ~~in~~at visible wavelengths could be reduced ~~by~~ ~~from~~by 5%–15% ~~when at~~with 1000 ng g$^{-1}$ BC ~~is present~~ in snow. Flanner et al. (2007) ~~developed~~ established a more comprehensive snow albedo model (the snow, ice, and aerosol radiation model; SNICAR) for a multilayer snowpack ~~using~~ based on the two-stream radiative transfer solution. In addition to BC, the SNICAR model also ~~accounts for~~considers the potential effects of dust particles and volcanic ash on snow albedo. Recently, ~~some~~ studies have indicated that the mixing state of BC and snow could effectively ~~change~~ alter snow albedo (Liou et al., 2011, 2014; Flanner et al., 2012; Liu et al., 2012; He et al., 2017, 2018a, b, c). Moreover, the snow grain shape ~~also has~~exerts an important influence on snow albedo (Kokhanovsky and Zege, 2004). Nonspherical snow grains ~~have~~ attain ~~weaker~~ a lower albedo reduction than that due to ~~snow~~ spher~~es~~ical snow grains (He et al., 2018c; Dang et al., 2016).

Although efforts have been made to optimize snow albedo models, current models still suffer from major limitations. Studies have ~~indicated~~ demonstrated that when BC in the atmosphere is coated with other aerosols, ~~it can significantly~~this greatly enhances light absorption via a lensing effect ~~compared with~~over uncoated BC, as investigated ~~using~~ via 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~~ persist for only a few hours after emission in ~~some~~ certain regions (Moteki et al., 2007; Moffet and Prather, 2009). Global aerosol models that simulate microphysical processes have ~~indicated~~ revealed that most BC is mixed with other particles within 1–5 days (Jacobson, 2001) at all altitudes (Aquila et al., 2011). However, ~~a problem~~it remains uncertain ~~is~~ ~~that~~ whether coated BC ~~is~~ ~~existed~~~~exists~~occurs in real snowpacks because the coating materials (e.g., salts and OC) ~~of coated~~other than BC may ~~be~~ dissolve~~d~~ during wet deposition. A recent study observing the individual particle structure and mixing states between ~~the~~ glaciers–snowpacks and the atmosphere based on field measurements and laboratory transmission electron ~~microscope~~microscopy (TEM) and energy dispersive X-ray

~~spectrometer~~spectrometry (EDX) instrument analysis (Dong et al., 2018) has ~~told~~ provided ~~the~~ an ~~truth~~answer. ~~They~~ It was found that ~~the~~ salt-coated BC was still observed in real snowpacks ~~in spite of~~ despite its lower proportion than that in the atmosphere due to the dissolution effect ~~within precipitating~~during snow precipitation. ~~For~~ Regarding OC, ~~that~~ the above study ~~didn't~~did not observe reduced OC components in LAPs. More notably, ~~that study further found~~it was also determined that the proportion of coated BC was even higher in snowpacks than that ~~that~~ in the atmosphere. All of the above observation results demonstrate~~d~~ that ~~the~~ coated BC particles ~~are existed~~ occur in real snowpacks and ~~were~~are even more common than ~~that~~those occurring in the atmosphere. Hence, the climate impacts of BC must be evaluated within the context of the BC coating effect ~~of the coating~~ on light absorption enhancement.

Although the BC coating effect on light absorption enhancement in the atmosphere ~~is~~ has been 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~~ thereby employing sulfate as ~~the~~ BC particle coating material with a constant absorption enhancement factor of ~1.5. Subsequently, Wang ~~at~~et al. (2017) ~~used~~ considered a similar constant light absorption enhancement factor ~~for~~ in their spectral albedo model for dirty snow (SAMDS). However, the above factor varies with the optical properties of different coating materials, ~~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~~notably controls light absorption enhancement. You et al. (2016) suggested that light absorption enhancement is highly correlated with visible or near-infrared (NIR) wavelengths and coating material. Furthermore, a core/shell Mie theory-based simulation study (Lack and Cappa, 2010) found that the attained light absorption enhancement was smaller for mildly absorbing coatings (e.g., OC) than ~~non-absorbing~~ that attained for nonabsorbing coatings (e.g., sulfate). Hence, ~~using~~ the use of a constant enhancement factor may ~~will~~ result in biased simulation estimates, ~~against refining our knowledge~~which prevents us from obtaining a better understanding of the hydrological and climate impacts of BC in snow ~~areas~~.

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

## 2 Methods

### 2.1 Modeling

### 2.1.1 Optical parameter calculations for snow coated in BC

Figure 1a and 1c show~~s~~ schematics of light absorption by externally and internally mixed particles (EMPs and IMPs, respectively). EMPs ~~refers to~~are particles ~~uncoated~~ not coated in BC mixed with other particles, while IMPs ~~refers to~~include BC, ~~that~~ which is assumed to be ~~a~~ the core material ~~coated~~ coating ~~by another~~ particles ~~acting~~ and acts as a shell (Kahnert et al., 2012). ~~For~~ Regarding ~~a~~ the nona~~non-a~~bsorbing shell, the overall light absorption includes contributions ~~from~~ of the BC core and ~~enhancement~~ absorption enhancement ~~from~~ due to ~~a~~ the lensing effect, while ~~for~~ regarding ~~an~~ the absorbing shell, the shell itself also contributes to light absorption. The lensing effect ~~means~~ indicates that when BC is coated with ~~the~~ a nona~~non-a~~bsorbing shell (or ~~the~~ an absorbing shell), the shell acts as a lens and focuses more photons onto the core than would reach it otherwise~~,~~ so that the light absorption effect ~~by~~ of the BC core ~~can be~~is enhanced (Bond et al., 2006).

To evaluate the BC coating effect on snow albedo, it is necessary to determine the optical parameters of ~~snow particles~~ coated ~~in~~ BC. The refractive index (RI) of BC was assumed to ~~be~~ ~~range from~~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 (nona~~non-a~~bsorbing and absorbing) were considered. The nona~~non-a~~bsorbing shell was represented ~~using~~ with sulfate, and its RI was ~~assumed~~ set to ~~range from~~be ~~be~~ $1.55–10^{-6}i$ following the atmospheric study of Bond et al. (2006). The absorbing shell was represented ~~using~~ with OC, which is a major light-absorbing particle in snow

(Wang et al., 2013). The RI of OC varies with the wavelength. Here, a fixed mass absorption coefficient (MAC) for OC of 0.3 m$^2$ g$^{-1}$ at 550 nm, a real RI value of 1.55, and a particle diameter of 200 nm were assumed, following the observations of Yang et al. (2009) and the study of Lack and Cappa (2010). The uncertainty ~~of~~ in snow albedo ~~of~~ considering the ~~coated~~ BC coating effect due to the OC MAC will be discussed in Section 3.4. Based on the Mie theory, an imaginary RI value of $-1.36 \times 10^{-2}$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~~ In regard to a core/shell-structured particle, the core and shell diameters refer to the BC core diameter and the ~~whole~~ whole-particle diameter, respectively. The BC diameter~~s are~~ usually ~~in the range of~~ranges from ~50–120 nm in the atmosphere (Corbin et al., 2018)~~,~~ and are typically larger by ~20 nm in snow due to ~~a~~ the removal process via wet deposition (Schwarz et al., 2013). Therefore, we assumed that the BC diameter in snow was 100 nm with a fixed monodisperse size distribution. The uncertainty ~~of~~ in snow albedo of ~~coated~~ the BC coating effect due to the BC size distribution will be ~~discussed~~ described in Section 3.4. The shell diameter was assumed to range from 110 ~~nm~~ to 300 nm based on Bond et al. (2006). ~~Core~~ The above core and shell diameters, RI, and wavelength were then ~~used~~ applied in a Mie model to derive the optical parameters of ~~the~~ core/shell particles, including the single scatter albedo (SSA), asymmetry factor ($g$), and extinction cross-section ($Q_{ext}$). The mass extinction coefficient (MEC) of ~~the~~ core/shell particles 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 a 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. The SNICAR model allows for a vertical multilayer distribution of snow properties, LAPs, and heating throughout the snowpack column. ~~Input~~ The input optical parameters (MEC, SSA, and *g*) of snow grains and BC were calculated ~~off-line~~offline ~~using~~ with the Mie theory. SNICAR provides snow albedo changes ~~from~~ due to uncoated and sulfate-coated BC particles~~on snow~~, ~~as well as~~in addition to dust particles and volcanic ash (for further details, ~~see~~ please refer to Flanner et al., 2007).

In this study, we assumed a ~~a~~ homogeneous semi-infinite snowpack and a solar zenith angle of 49.5°, whose cosine value (0.65) represents the insolation-weighted mean solar zenith cosine ~~for~~ in the sunlit Earth hemisphere (Dang et al., 2015). The snow grain optical effective radius was varied from 50 to 1000 μm (~~with a~~at 50-μm intervals) to characterize snow aging. ~~Meanwhile~~Moreover, the BC concentration~~s~~ ~~were~~ was assumed to ~~be in the range of~~ range from 0-1000 ng g$^{-1}$ (~~with a~~at 10-ng g$^{-1}$ intervals) to ~~demonstrate~~ simulate clear to polluted snow, which was based on ~~the~~ global field observations ~~with~~ of the BC concentration~~s~~ in snowpacks mostly below 1000 ng g$^{-1}$ (e.g., Doherty et al., 2010, 2014; Wang et al., 2013; Li et al., 2017, 2018; Pu et al., 2017; Zhang et al., 2017, 2018). These parameters ~~are~~ were also applied ~~for~~ in the subsequent parameterizations (~~see~~ please refer to Section 2.3). In addition, we note that the SNICAR model ~~used~~ adopted in this study ~~was~~ is the default version ~~that assumes~~assuming BC-snow external mixing and ~~snow~~ spher~~es~~ical snow grains (Flanner et al., 2007). Although the mixing state of BC and snow grains~~,~~ and the snow grain shape ~~can~~ affect the snow albedo, ~~the~~ empirical parameterizations ~~for~~ describing the effect of BC internally mixed with snow grains on snow albedo ~~has been~~were developed by He et al. (2018c), and the albedo of a snowpack consisting of nonspherical snow grains ~~can be~~was ~~mimicked~~ simulated ~~by using a~~with smaller spherical grains ~~of spherical shape~~ (Dang et al. 2016). Therefore, users ~~can~~ may combine the empirical parameterizations developed by He et al. (2018c) and Dang et al. (2016) with ~~the~~ our empirical parameterizations ~~by us~~ (~~see~~ please refer to Section 2.3) to study the effect of the internal mixing of BC with snow grains, snow grain shape, and coated BC particles on snow albedo.

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

### 2.2 Calculation of the broadband snow albedo

The spectral albedo ($\alpha_\lambda$) was integrated over the solar spectrum ($\lambda = 300$–$2500$ nm) and weighted ~~by~~ based on the incoming solar irradiance ($S_\lambda$) to calculate the broadband snow albedo ($\alpha_{\text{integrated}}$):

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

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

### 2.3 Parameterizations

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

$$E_{\alpha,\text{integrated}} = \frac{\alpha_{int,\text{integrated}}}{\alpha_{ext,\text{integrated}}} \qquad (2)$$

where $\alpha_{ext,\text{integrated}}$ and $\alpha_{int,\text{integrated}}$ are the broadband snow albedos for EMPs and IMPs, respectively. Following a previous empirical formulation (Hadley and Kirchstetter, 2012), $E_{\alpha,\text{integrated}}$ was parameterized as:

$$E_{\alpha,\text{integrated,para}} = a_0 \times (C_{BC})^{a_1} + a_2 \qquad (3)$$

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

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

### 2.4 Calculation of the in situ snow albedo and radiative forcing

In situ broadband clear-sky ($\alpha_{\text{integrated}}^{\text{clear,in-situ}}$) and cloudy-sky ($\alpha_{\text{integrated}}^{\text{cloudy,in-situ}}$) albedos were separately calculated ~~separately~~ based on corresponding in situ snow-LAP parameters and SBDART simulated clear-~~sky~~ and cloudy-sky incoming solar irradiance levels, respectively. We assumed a ~~a~~ semi-infinite snowpack due to the limited available snow depth measurements. The BC and OC concentrations were collected from in situ field measurements (e.g., Doherty et al., 2010, 2014; Wang et al., 2013; Li et al., 2017, 2018; Pu et al., 2017; Zhang et al., 2017, 2018). ~~The~~A snow grain radius of 100 (1000) µm was assumed for fresh (old) snow, which is comparable to previous observations at mid-~~to~~high latitudes in winter (Wang et al., 2017; Shi et al., 2020). The value of the solar zenith angle was calculated based on the longitude, latitude, and sampling time at each sampling site. The in situ all-sky albedo ($\alpha_{\text{integrated}}^{\text{all−sky,in−situ}}$) was then calculated ~~using~~ based on weighted clear-~~sky~~ and cloudy-sky albedo values depending on the cloud fraction (CF), given as:

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

~~In~~ The in situ radiative forcing ~~by~~ due to LAPs was calculated by multiplying the derived broadband albedo reduction by the downward shortwave flux at the snow surface (Dang et al., 2017). We ~~point out~~ note that the radiative forcing was calculated ~~using~~ with the January-February average solar radiation ~~for~~ in NA and NC~~, while~~ and the April-May average solar radiation ~~for~~ in the Arctic and TP according to the periods of corresponding ~~filed~~field campaigns. In this study, we mainly estimated the relative impacts of internal ~~mixing to~~and external mixing on snow albedo and radiative forcing, which ~~is~~ are hence not influenced by the chosen solar radiation level. Figure S2 shows ~~the~~ spatial distributions of the 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

Figure 1b and 1d show~~s~~ the light absorption enhancement~~,~~ and $E_{abs}$, respectively, ~~for~~ due to coated BC particles. $E_{abs}$ is defined as the ratio of the light absorption ~~for~~ due to coated ($LA_{int}$) ~~versus~~ and uncoated BC particles ($LA_{ext}$)~~,~~ ($E_{abs} = \frac{LA_{int}}{LA_{ext}}$). Based on

Bond et al. (2006), we show the most common core/shell ratios (the ratio of the diameter of the whole particle to that of 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~~ considered detailed core/shell ratios ranging from 1.1 to 3.0 (~~in~~ at intervals of 0.1) ~~for~~ in the parameterizations (see Section 3.5). $E_{abs}$ varies with the wavelength and increases with the core/shell ratio, in contrast to the default $E_{abs}$ value ~~used~~ employed in the original SNICAR model, which remains constant. ~~For~~ Regarding ~~a non-absorbing~~nonabsorbing shells, the light absorption of IMPs is ~~larger~~ higher than– that of EMPs across all wavelengths (300–1400 nm). ~~For~~ Regarding ~~an~~ absorbing shells, $E_{abs}$ is similar to ~~the~~ that of nona~~non-a~~bsorbing shells in the NIR~~,~~ range but decreases~~becomes smaller~~ in the visible (VIS) light and ~~ultra-violet~~ultraviolet (UV) light ranges, which ~~implies~~ indicates that ~~the~~ absorbing shells reduce~~s~~ whole-particle light absorption and ~~contributes~~ negatively contribute to $E_{abs}$. This ~~is~~ occurs because compared ~~with~~ to ~~non-absorbing~~the nonabsorbing shell, the absorbing shell, although it absorbs additional incident photons,~~ but~~ causes fewer photons to reach the core, so that the photons absorbed by the lensing effect and ~~the~~ BC core ~~will be~~are reduced. In ~~such a~~this case, the number of additional photons absorbed by the shell ~~are~~ is ~~fewer~~ smaller than the ~~reduced~~ number of fewer photons absorbed by the lensing effect and ~~the~~ BC core, causing ~~that~~ the total absorption ~~by~~ of absorbing shell-coated BC particles ~~will~~to be ~~smaller~~ lower than that of nona~~non-a~~bsorbing shell-coated BC particles (Lack and Cappa, 2010). Furthermore, the absorbing shell reduces $E_{abs}$ to <1 in the UV range at high core/shell ratios, ~~implying~~ suggesting that the lensing effect on absorption at ~~those~~ these wavelengths ~~cannot~~ does not ~~recover~~ match the BC core absorption reduction, resulting in fewer photons reaching the core, which is similar to the results reported by Lack and Cappa (2010).

### **3.2 Impact on the spectral snow single-scattering properties and albedo~~s~~**

In a real snowpack, BC ~~can~~ effectively enhances the snow single-scattering co-albedo (1–ω), but its effect on other snow optical parameters, such as the asymmetry factor and extinction efficiency, is negligible (He et al., 2017). Therefore, we focus our discussion on the coating-induced enhancement of the snow single-scattering co-albedo ($E_{1-\omega}$), snow albedo ($E_\alpha$), and snow albedo reduction ($E_{\Delta\alpha}$). ~~The~~ $E_{1-\omega}$ is defined as the ratio of the snow single-scattering co-albedo ~~with~~ due to coated BC particles ($1-\omega_{int}$) ~~versus~~ to that ~~with~~ due to uncoated BC particles ($1-\omega_{ext}$)~~,~~ ($E_{1-\omega}=\frac{1-\omega_{int}}{1-\omega_{ext}}$). Similar definitions were ~~used~~ adopted for $E_\alpha$ ($E_\alpha=\frac{\alpha_{int}}{\alpha_{ext}}$) and $E_{\Delta\alpha}$ ($E_{\Delta\alpha}=\frac{\Delta\alpha_{int}}{\Delta\alpha_{ext}}$), where $\alpha_{int}$ and $\alpha_{ext}$ are the snow albedo values ~~with~~ due to coated and uncoated BC particles, respectively, and $\Delta\alpha_{int}$ and $\Delta\alpha_{ext}$ are the snow albedo reductions due to coated and uncoated BC particles, respectively.

Figure 2 shows the ~~varied~~ variation in 1–ω and $E_{1-\omega}$ depending on ~~different~~ the BC concentration~~s~~, core/shell ratio~~s~~, and coating material~~s~~. ~~For~~ Regarding either athe nona~~non-a~~bsorbing ~~shell~~ or absorbing shell, $1-\omega_{int}$ is usually ~~larger~~ higher than $1-\omega_{ext}$ in the VIS range, while the coating effect ~~has~~ exerts little ~~impacts at wavelength~~impact at wavelengths > 1200 nm~~, which is due to that~~ because the optical properties of snow ~~is~~are ~~effectively~~ mainly affected by LAPs in the VIS~~,~~ range but primarily by snow itself at ~~wavelength~~wavelengths > 1200 nm. In addition, $E_{1-ω}$ increases with ~~increased~~increasing core/shell ratio~~s~~, and the wavelength ~~of~~ with the maximum $E_{1-ω}$ value ~~is dependent~~depends on the BC concentration~~s~~ and core/shell ratio~~s~~. Moreover, the absorbing shell ~~shows~~ reveals a negative impact ~~for~~on $E_{1-ω}$ ~~compared with~~over the nona~~non-a~~bsorbing shell, especially in the UV range.

Snow albedo is ~~effectively~~ notably ~~affected~~ influenced by various factors, such as the snow grain size, LAP content, and solar zenith angle, which has been widely ~~discussed~~ examined and verified through model simulations and experimental measurements ~~by~~ in previous studies (e.g., Warren and Wiscombe, 1980; Hadley and Kirchstetter, 2012; Wang et al., 2017). In this study, we mainly focus on the BC coating effect ~~of BC~~ on snow albedo. Figure 3 shows the spectral snow albedo values ~~for~~ due to coated ($α_{int}$) and uncoated BC particles ($α_{ext}$)~~,~~ and the ratios ($E_α$) of $α_{int}$ ~~versus~~ to $α_{ext}$. ~~In consistent~~ Consistent with 1–ω, the impact of the coating effect on snow albedo is mainly ~~presents at wavelengthpresent~~observed at wavelengths < ~1200 nm (Figures 3a versus 3b, and Figures 3d versus 3e), where the higher the BC concentration is (or the ~~larger~~ higher the core/shell ratio is), the larger the difference ~~of~~in snow albedo~~s~~ between uncoated and coated BC particles~~is~~. Hadley and Kirchstetter (2012) also found a ~~smaller~~ lower snow albedo ~~for~~ due to internally mixed particles ~~relative to~~than that ~~for~~ due to externally mixed particles. This phenomenon is also obvious ~~on~~for $E_α$, which decreases with ~~increased~~ increasing BC concentration and core/shell ratio~~s~~ in the VIS and NIR ranges (Figure 3c and 3f, respectively). ~~For~~ At a given BC concentration and core/shell ratio, $E_{\alpha}$ generally decreases with the wavelength from the UV range to the VIS~~,~~ range and then increases from the VIS range to the NIR range, which ~~is corresponding~~ corresponds to the ~~results of~~ $E_{abs}$ and $E_{1-\omega}$ results. ~~On the other hand~~In contrast, the $E_{\alpha}$ ~~values~~ –values ~~for~~ considering the nona~~non-a~~bsorbing and absorbing ~~shell is~~shells are comparable~~ with each other at wavelength~~ at wavelengths > ~800 nm. However, when the wavelength < ~800 nm, $E_{\alpha}$ ~~for~~ considering the absorbing shell is ~~larger~~ higher than that ~~for~~ considering ~~non-absorbing~~the nonabsorbing shell, and the difference increases with ~~the decreased~~ decreasing wavelength and ~~increased~~increasing core/shell ratio. Moreover, ~~for~~ regarding the absorbing ~~shell~~shells, the snow albedo ~~for~~ due to coated BC particles is higher than that ~~for~~ due to uncoated BC particles at < ~350 nm at ~~large~~ high core/shell ratios~~, which are due to that~~ because the light absorption ~~by~~ of internally mixed particles ~~for~~ with absorbing ~~shell~~shells is ~~smaller~~ lower than that ~~by~~ of externally mixed particles at ~~those~~ the same wavelengths, as previously ~~discussed~~ described in Section 3.1. These results indicate that the material of the particle shell also plays an important role ~~for~~in snow albedo in the UV and VIS ranges. We note that the solar radiative flux is very ~~small~~ low at wavelengths < 350 nm, so that the coating effect at ~~those~~ these wavelengths may ~~have~~ contribute little ~~contributionscontribution~~ to the total light absorption and broadband snow albedo~~s,~~ but ~~which~~ may potentially influence the photochemical reactions in a snowpack (Grannas et al., 2007).

Furthermore, Figure 4 shows the spectral snow albedo reduction~~s~~ caused by coated ($\Delta\alpha_{int}$) and uncoated BC particles ($\Delta\alpha_{ext}$)~~,~~ and the ratio~~s~~ ($E_{\Delta\alpha}$) of $\Delta\alpha_{int}$ ~~versus~~ to

$\Delta\alpha_{ext}$. Generally, $\Delta\alpha_{int}$ is larger than $\Delta\alpha_{ext}$, and the core/shell ratio dominates the variation~~s~~ ~~of~~in $E_{\Delta\alpha}$ across ~~the~~ all wavelengths ~~of~~ from 300-1400 nm, while the impact of the BC content is mainly ~~focuses~~ manifested ~~on~~ from 500-1000 nm. ~~In consistent~~ Consistent with $E_{1-\omega}$ and $E_{\alpha}$, the impact of the material of the particle shell is negligible at a wavelength > ~800 nm, but ~~the~~ $E_{\Delta\alpha}$ for the absorbing shell is ~~smaller~~ lower than that for ~~non-absorbing~~ the nonabsorbing shell at a wavelength < ~800 nm. Moreover, ~~the~~ $E_{\Delta\alpha}$ is < 1 for the absorbing shell at ~~wavelengthat~~ wavelengths < ~350 nm ~~at~~ and ~~large~~ high core/shell ratios. It is ~~interesting~~ noteworthy that the coating effect still ~~has~~ yields an obvious impact on snow albedo reduction at ~~wavelength~~wavelengths > ~1200 nm, which is different ~~with~~from $E_{1-\omega}$ and $E_{\alpha}$.

### 3.3 Impact on the broadband snow single-scattering properties and albedo~~s~~

Compared ~~with~~ to the spectral optical properties, our broadband results have wider implications for the research community. Figure 5 shows the spectrally weighted 1-ω ~~for~~ due to coated ($1-\omega_{int,\ integrated}$) and uncoated BC particles ($1-\omega_{ext,\ integrated}$)~~,~~ and the ~~raitos~~ ratio ($E_{1-\omega,\ integrated}$) of $1-\omega_{int,\ integrated}$ ~~versus~~ to $1-\omega_{ext,\ integrated}$. In general, $1-\omega_{int,\ integrated}$ is larger than $1-\omega_{ext,\ integrated}$, and $E_{1-\omega,\ integrated}$ ~~increased~~ increases with the BC concentration and core/shell ratio but is ~~, but is~~ little affected by the snow grain size. $E_{1-\omega,\ integrated}$ ~~is~~ ~~in a range of~~ranges from 1.0 to ~1.35 and 1.0 to ~1.23 for the nona~~non-a~~bsorbing and absorbing ~~shell~~shells, respectively, with the BC concentration~~s~~ ~~within~~ lower than 1000 ng $g^{-1}$ ~~and~~ at core/shell ratios ~~of~~ ranging from 1.2-2.5. ~~For~~ At a given BC concentration and core/shell ratio, $E_{1-\omega}$~~,~~ integrated ~~for~~ considering ~~non-absorbinga~~the nonabsorbing shell is ~~larger~~ higher than that ~~for~~ considering ~~an~~the absorbing shell. In addition, $E_{1\text{-}\omega,\ \text{integrated}}$ determined ~~of~~with the original SNICAR model, is ~~closed~~close to that ~~of~~ considering the nona~~non-a~~bsorbing shell at a core/shell ratio of 1.5.

Figure 6 shows the spectrally weighted snow albedo ~~for~~ due to coated ($\alpha_{\text{int, integrated}}$) and uncoated BC particles ($\alpha_{\text{ext, integrated}}$)~~,~~ and the ratio~~s~~ ($E_{\alpha,\ \text{integrated}}$) of $\alpha_{\text{int, integrated}}$ ~~versus~~ to $\alpha_{\text{ext, integrated}}$. Generally, $\alpha_{\text{int, integrated}}$ is ~~smaller~~ lower than $\alpha_{\text{ext, integrated}}$ by 0 to 0.069 (0 to 0.051), and $E_{\alpha,\ \text{integrated}}$ ~~varies~~ ranges from 1 to ~0.903 (1 to ~0.924) ~~for~~ considering the nona~~non-a~~bsorbing (absorbing) shell ~~with~~ at BC concentrations from 0 to 1000 ng $g^{-1}$, with the snow grain radius ranging from 100 ~~μm~~ to 500 μm, and the core/shell ratio~~s~~ ranging from 1.2 to 2.5. $E_{\alpha,\ \text{integrated}}$ ~~shows~~ exhibits a ~~decreased~~decreasing trend with ~~increased~~ increasing BC concentration, core/shell ratio and snow grain size. In addition, the difference between $\alpha_{\text{ext, integrated}}$ and $\alpha_{\text{int, integrated}}$ (~~-~~or $E_{\alpha,\ \text{integrated}}$) for ~~a~~the nona~~non-a~~bsorbing shell is larger (~~-~~or smaller) than that for ~~an~~the absorbing shell. If considering these coating effects in real environments~~. For example~~, e.g., in clean snow, such as in North ~~American~~America ~~with~~ at a typical BC concentration of ~50 ng $g^{-1}$ (Doherty et al., 2014), the difference between $\alpha_{\text{ext, integrated}}$ and $\alpha_{\text{int, integrated}}$ ~~is in a range of~~ranges from 0.002-0.017 and 0.001-0.012 ~~for~~ considering the ~~non-absorbing~~nonabsorbing and absorbing ~~shell~~shells, respectively, ~~with~~ at core/shell ratios ~~of~~ from 1.2-2.5 and snow grain ~~radius~~radii ~~of~~ from 100-500 μm. In contrast, in polluted snow, such as in ~~Northeastern~~northeastern China, the BC concentration is typically ~1000 ng $g^{-1}$ in industrial regions. ~~the~~The difference between $\alpha_{\text{ext, integrated}}$ and $\alpha_{\text{int, integrated}}$ ranges from

0.008-0.069 and 0.007-0.051 ~~for~~ considering the nona~~non-a~~bsorbing and absorbing ~~shell~~shells, respectively. These results ~~show~~ indicate that the impact of the coating effect on snow albedo ~~can leads the~~may lead to a reduction in snow albedo ~~reduced~~ by ~2% in clean snow and ~10% in polluted snow ~~for~~ due to coated BC ~~than that for~~ particles ~~compared with~~below the snow albedo due to uncoated BC particles. In addition, the sensitivity of $E_{\alpha}$~~,~~ $_{\text{integrated}}$ ~~towith~~to BC decreases with increasing BC concentration due to the nonlinear effect of BC on snow albedo (Flanner et al., 2007).

Figure 7 shows the spectrally weighted snow albedo reduction~~s~~ ~~by~~ due to coated ($\Delta\alpha_{\text{int, integrated}}$) and uncoated BC particles ($\Delta\alpha_{\text{ext, integrated}}$)~~,~~ and the ratio~~s~~ ($E_{\Delta\alpha,\ \text{integrated}}$) of $\Delta\alpha_{\text{int, integrated}}$ ~~versus~~ to $\Delta\alpha_{\text{ext, integrated}}$. ~~Different from~~In contrast to $E_{\alpha,\ \text{integrated}}$, $E_{\Delta\alpha,\ \text{integrated}}$ is dominated by the core/shell ratio~~,~~ but ~~little dependent~~slightly depends on the snow grain size (Figures 7c and 6f, respectively). In addition, $E_{\Delta\alpha,\ \text{integrated}}$ ~~presentspresented~~exhibits a slight ~~decreased~~decreasing trend with ~~increased~~increasing BC concentration. Comparing Figure 7c and f, we find that the~~ material of~~ particle shell ~~presents~~material ~~has~~exerts a distinct integrated impact on $E_{\Delta\alpha}$~~, integrated~~. $E_{\Delta\alpha,\ \text{integrated}}$ mostly ~~falls in athe range of~~ranges from 1.11 to ~1.80 (1.10 to ~1.33) for the nona~~non-a~~bsorbing (absorbing) ~~shell~~shells ~~with~~ at core/shell ratios from 1.2 to 2.5. Our results ~~were~~ are comparable ~~with~~ to ~~the~~those of a previous study~~ that~~ in which the snow albedo reduction ~~of~~ due to ~~BC snow~~BC/snow internal mixing ~~is~~was larger than that ~~of~~due to external mixing by a factor of 0.2-1.0 (He et al., 2018c). ~~On the other hand~~However, the $E_{\Delta\alpha}$~~,~~ $_{\text{integrated}}$ value retrieved from the original SNICAR model ~~shows~~demonstrates only ~~shows~~ a small variation ~~of~~ from 1.23–1.31. This is similar to ~~a~~ the nona~~non-a~~bsorbing shell ~~with~~ at a core/shell ratio of ~1.5, which ~~implies~~ suggests that the original SNICAR model only reflects ~~a~~ the coating effect on snow albedo reduction at an intermediate core/shell ratio, ~~and~~ which may lead to possible biases ~~of~~ ranging from -10% to 50% in ~~the~~ snow albedo reduction calculations.

### 3.4 Uncertainties

Although the imaginary RI value of OC has been theoretically calculated (Section 2.1), we note that in a real snowpack, there ~~is~~ exists a ~~large~~ high uncertainty because the types and optical properties of OC ~~varies~~vary 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 the imaginary RI value on the $E_{\alpha,\ \mathrm{integrated}}$, and $E_{\Delta\alpha,\ \mathrm{integrated}}$ values by increasing and decreasing the calculated imaginary RI value by 50% (Figure S1), which studies have ~~shown~~ revealed to be plausible (e.g., Lack et al., 2012). We ~~find~~ found the imaginary RI uncertainty to be ±1% for $E_{\alpha,\ \mathrm{integrated}}$ and ±5% for $E_{\Delta\alpha,\ \mathrm{integrated}}$.

In addition, observations ~~show~~ have demonstrated large ~~variation~~variations in the size distribution of atmospheric and snowpack BC particles (Schwarz et al., 2013), which ~~can~~ may affect the snow optical properties and albedo (He ~~at~~et al., 2018b). Therefore, we examined the effects of the BC particle size on $E_{\alpha,\ \mathrm{integrated}}$ and $E_{\Delta\alpha,\ \mathrm{integrated}}$ with two additional BC particle diameters of 50 ~~nm~~ and 150 nm, which ~~are~~ occur within the observed size ranges (Schwarz et al., 2013) and are comparable to the BC particle sizes ~~used~~ adopted in other studies (e.g., He et al., 2018b). We find that the uncertainty attributed to the BC particle diameter is ±1% for $E_{\alpha,\ \mathrm{integrated}}$ and ±13% for $E_{\Delta\alpha,\ \mathrm{integrated}}$. According to Equation 2, the uncertainty ~~for~~ in $E_{\alpha,\ \mathrm{integrated}}$, is equivalent to that ~~for~~ in the snow albedo, and the uncertainty ~~for~~ in $E_{\Delta\alpha,\ \mathrm{integrated}}$, is equivalent to that ~~for~~ in the snow albedo reduction. Therefore, the total uncertainty related to the imaginary RI value and BC particle diameter is ±1.4% for $E_{\alpha,\ \mathrm{integrated}}$ (snow albedo) and ±13.9% for $E_{\Delta\alpha,\ \mathrm{integrated}}$ (snow albedo reduction).

Another important issue is that in real environments, BC mixtures ~~with~~ containing other species are likely much more complex than are uniform coatings on spheres~~,~~.~~;~~ Hence~~hence~~, a core-shell assumption seems somewhat dubious. However, a recent study observing the individual particle structure and mixing states between ~~the~~ glaciers–snowpacks and the atmosphere (Dong et al., 2018) has found that fresh BC particles are generally characterized ~~with~~by a fractal morphology, which ~~has a large quantity~~abundantly occur in the atmosphere. In contrast, in a ~~the~~ snowpack, aged BC particles dominated the BC content, and the mixing states of aged BC particles ~~changechanged~~ largely changed to ~~the~~ internal mixing forms with BC ~~as~~ at the core. This process ~~is~~ was characterized by the initial transformation from a fractal structure to a spherical morphology and the subsequent growth of fully compact particles during the transport and deposition process. Therefore, a core-shell assumption for coated BC particles in a snowpack seems to be plausible. In addition, most ~~filed~~field measurements ~~can not cannot~~have not captured the explicit structure of coated BC particles due to the limited observation methods (e.g., Doherty et al., 2010, 2014; Wang et al., 2013; Li et al., 2017, 2018; Pu et al., 2017; Zhang et al., 2017, 2018)~~,~~; therefore, even if a model ~~for~~ of the explicit BC ~~structure~~structures was developed, researchers ~~are hard to use it for~~ ~~have~~experience difficult~~y~~ies studying the effect of coated BC particles on snow albedo reduction~~s~~ at present. Moreover, a core-shell assumption for coated BC particles in the atmosphere has been widely applied ~~by~~ in most global climate models (e.g., Jacobson, 2001; Bond et al. 2013), so ~~that~~ our parameterizations ~~for~~ describing coated BC particles in a snowpack ~~can~~ may be easily linked to ~~them~~these models. In summary, we indicate that a core-shell assumption ~~for~~ describing coated BC particles in a snowpack is plausible and practical for field observations and model simulations at present ~~in~~ despite ~~of the~~ possible uncertainties. However, with the ~~developments~~development of measurement methods and climate models, ~~building~~ a more explicit structure ~~for~~ characterizing coated BC particles in a snowpack is actually needed in the future.

### 3.5 Parameterizations of the coating effect

Figure 8 compares parameterized $E_{\alpha,\ \mathrm{integrated,\ para}}$ values ~~with~~ to SNICAR-modeled $E_{\alpha,\ \mathrm{integrated}}$ values, and Tables S1 and S2, respectively, list the empirical coefficients (~~see~~ please refer to Section 2.3) derived from the nonlinear regression process~~es~~. This parameterization is applicable under the assumptions of a semi-infinite snowpack, BC-snow external mixing, and spherical snow grains, as mentioned in Section 2. Generally, $E_{\alpha,\ \mathrm{integrated,\ para}}$ and $E_{\alpha,\ \mathrm{integrated}}$ ~~show~~ exhibit a strong correlation, with $R^2 = 0.988$ (0.986) for ~~a~~ the nona~~non-a~~bsorbing shell and $R^2 = 0.987$ (0.986) for ~~an~~ the absorbing shell in relatively clean (polluted) snow, and root mean squared errors of $1.81 \times 10^{-3}$ (4.70 × ~~-~~$10^{-3}$) and $1.41 \times 10^{-3}$ ($3.76 \times 10^{-3}$), respectively. ~~Biases~~ The biases ~~for~~ in $E_{\alpha,\text{ integrated, para}}$ are ~~smallest~~ the lowest ~~for~~ at intermediate BC concentrations~~,~~ but become relatively ~~larger~~ high at extremely low or high ~~values~~concentrations, mainly due ~~mainly~~ to processes within the nonlinear regression method. In addition, the snow grain size ~~has~~ exerts a ~~small~~ limited ~~impacts~~impact on the accuracy of the parameterized results~~,~~ so that these parameterizations can be applied ~~in~~ to either fresh ~~snow~~ or old snow types. Overall, the integrated $E_{\alpha}$~~, integrated~~ value ~~can be~~is ~~well~~ suitably reproduced by $E_{\alpha,\text{ integrated, para}}$, and the parameterizations are applicable ~~in~~ under various snow pollution conditions ~~with~~ at BC concentrations ranging from 0-1000 ng g$^{-1}$, core/shell ratios ranging from 1.1 to 3.0, and different coating materials (nona~~non-a~~bsorbing and absorbing ~~shell~~shells). We note that if the BC concentration is ~~larger~~ higher than 1000 ng g$^{-1}$, the parameterization ~~for~~ describing relatively polluted snow is also applicable with a ~~small~~ low negative bias.

Therefore, ~~other~~ future studies ~~can~~ may estimate the BC coating effect ~~of BC~~ on snow albedo~~s~~ and radiative forcing very conveniently by combining the original SNICAR model or other snow radiative forcing ~~model~~models with our new parameterizations, which may reduce the snow albedo simulation bias. ~~On the other hand~~However, although most global climate models (GCMs) account for coated BC particles in the atmosphere, they barely consider the BC coating effect ~~for BC~~ in snow

~~areas~~ (Bond et al., 2013). In addition, different GCMs apply ~~different~~ varying types of snow radiative transfer models, which ~~means~~ indicates that one physical mechanism ~~responsible for~~describing the BC coating effect in snow ~~areas~~ ~~cannot~~ may not be suitable for all GCMs. Hence, our parameterizations are ~~good~~ suitable for ~~the~~ climate models ~~to have~~and provide an option ~~for~~ to capture BC coating effects in snow ~~areas~~.

### 3.6 Measurement-based estimate of the coating effect

To evaluate the BC coating effect ~~of BC~~ on both the snow albedo and radiative forcing in a real snowpack, we collected in situ measurements of BC and OC concentrations in snow (Figure 9) during field campaigns in the Arctic in the spring from 2007–2009 (Doherty et al., 2010), in North America ~~in~~ from January–March 2013 (Doherty et al., 2014), in northern China ~~in~~ from January–February 2010 and 2012 (Ye et al., 2012; Wang et al., 2013), and on the Tibetan Plateau in the spring of 2010 and 2012 (Wang et al., 2013; Li et al., 2017, 2018; Pu et al., 2017; Zhang et al., 2017, 2018). ~~Measurements~~ The measurements ~~are~~ were separated into four geographical regions (Figure 9c): the Arctic, North America (NA), northern China (NC), and the Tibetan Plateau (TP). An absorbing shell consisting of OC was assumed in the measured snowpack data, which is plausible because previous studies have found that OC is the dominant coating in the atmosphere (e.g., Cappa et al., 2012) and snow (Dong et al., 2018). The OC/BC mass ratio ~~is~~ generally ranges from 1 to 10, with the corresponding core/shell ratio ranging from 1.3 to 2.5 (Figure 9b). The average core/shell ratio was the highest (2.45) ~~in~~on the TP, followed by values of 1.92 and 1.81 in the Arctic and

NC, respectively, and was the lowest (1.31) in NA (Figure 9d). These results reveal that the BC coating effect ~~had~~ exerted a larger impact on snow albedo ~~in~~ on the TP than that in the other regions. In this study, the assumption that all measured OC ~~resides~~ occurs as a coating on BC particles ~~were~~was mainly ~~used~~ adopted to ~~show~~ reveal the upper bound of the coating effect on snow albedo reduction, which is comparable ~~with~~ to ~~the~~ previous studies (e.g., He et al. 2018c).

Figure 10 shows ~~the~~ statistics ~~for~~ of the snow albedo reduction~~s~~ and radiative forcing in the different regions for fresh snow (snow grain radius =100 μm) and old snow (snow grain radius =1000 μm). ~~The s~~Spatial distributions of the attained snow albedo reduction~~s~~ and radiative forcing are ~~presented~~ shown in ~~Figure~~Figures S3 and S4, respectively. Briefly, the TP snowpack suffers the highest snow albedo reduction (0.066), and the regional average snow albedo reduction~~s~~ ~~are~~ is lower in NC (0.055), NA (0.009), and the Arctic (0.007) for fresh snow in the case of external mixing (Figure 10a). Accordingly, the regional average radiative forcing is 11.63, 4.42, 0.97, and 0.56 W m$^{-2}$ ~~in~~ on the TP, NC, NA, and the Arctic, respectively (Figure 10b). In the case of internal mixing, the regional average snow albedo reduction~~s~~ ~~are~~ is 0.084, 0.065, 0.011, and 0.009 ~~in~~ on the TP, NC, NA, and the Arctic, respectively, with ~~the~~ corresponding radiative ~~forcing~~forcings of 14.84, 5.51, 1.11, and 0.69 W m$^{-2}$~~.~~, respectively. Figure 11 shows ~~the~~ a comparison~~s~~ of internal mixing to external mixing. ~~For~~ In fresh snow, we find that coated BC particles result~~s~~ in greater snow albedo reductions ~~compared~~ than those ~~with~~ due to uncoated BC particles by factors of 1.27, 1.19, 1.13, and 1.23 ~~in~~ on the TP, NC, NA, and the Arctic, respectively (Figures 11a and 11b, respectively). Correspondingly, we find that the coating effect ~~leads~~ yields radiative forcing ~~by~~ values of 1.27, 1.20, 1.14, and 1.22, respectively, ~~for~~ in these ~~same~~ regions. The ~~highest~~ largest (~~lowest~~smallest) enhancement was found ~~in~~on the TP (NA), which corresponds to the highest (lowest) OC/BC mass ratio and core/shell ratio ~~in~~on the TP (NA). ~~For~~ In regard to old snow, the regional average snow albedo reduction~~s~~ ~~are~~ is 0.17 (0.21), 0.14 (0.17), 0.028 (0.033) and 0.022 (0.027) ~~in~~ on the TP, NC, NA, and the Arctic, respectively, for external (internal) mixing (Figure 10c). The corresponding radiative ~~forcing~~forcings are 38.2 (47.6), 19.2 (22.7), 4.6 (5.2) and 3.6 (4.6) W m$^{-2}$, respectively (Figure 10d). The enhancement of snow albedo reduction~~s~~ due to the BC coating effect ~~are~~is 1.24, 1.15, 1.13, and 1.23 ~~in~~ on the TP, NC, NA, and the Arctic, respectively (Figure 11c). The corresponding radiative forcing reduction~~s~~ ~~are~~ is 1.24, 1.16, 1.14, and 1.22, respectively (Figure 11d). The enhancement ~~shows~~ exhibits a slight decrease with snowpack aging, which is consistent with the results shown in Figure 7. ~~Of note~~Notably, we found that the contribution of the coating effect to light absorption ~~has exceeded~~ exceeded that of dust over most areas of northern China after ~~comparing~~comparison ~~with~~ to previous studies of dust in snow (Wang et al., 2013, 2017; Pu et al., 2017), which further demonstrated the critical role of the BC coating effect~~s~~ in snow albedo evaluation.

In contrast to previous studies, we note that an enhanced light absorption in snow ~~areas~~ ~~from~~ due to the BC coating effect should be ~~taken into account~~considered, especially in the Arctic and ~~the~~ TP. Arctic sea ice has ~~shown a sharp decline~~sharply declined 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~~Multimodel ensemble simulations ~~indicate~~ have indicated 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 the 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~~ freshwater supplies could disappear by the middle of the 21st century. Observed evidence ~~suggests~~ has suggested that BC deposition is a ~~significant~~ major contributing factor to this retreat (Xu et al., 2009), but the quantitative~~ly~~ modeling of the BC effect ~~of BC~~ on glacier dynamics is a challenge, partly because of the incomplete radiative transfer mechanisms within current models. Due to the ~~significant~~ notable contribution of BC to ~~retreating~~ the retreat in Arctic sea ice and Tibetan glaciers~~,~~ and the strong enhancement of light absorption ~~by~~ due to coated BC particles, the coating effect must now be considered in climate models that are designed to accurately reconstruct both ~~the~~ historical ~~record~~records and future ~~change~~changes.

## 4 Conclusions

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

Parameterizations ~~for~~ describing the coating effect ~~are~~ were further developed for ~~use~~ application in snow albedo and climate models. ~~Parameterized~~ The parameterized and simulated results ~~show~~ exhibit strong correlations ~~for~~ in both clean and polluted snowpacks. The root mean squared error of the parameterized $E_{\alpha,\ \mathrm{integrated,\ para}}$ values is ~~low~~ small ($1.41 \times 10^{-3}$). A list of empirical coefficients for parameterizations ~~is~~ was provided suitable for most seasonal snowpack field cases, with BC concentrations ~~within~~ lower than 1000 ng g$^{-1}$, snow grain sizes ranging from 50–1000 µm, and core/shell ratios ranging from 1.1 to 3.0. We demonstrated that these parameterizations ~~can~~ could reduce the simulation bias ~~for~~ regarding local experiments in snow albedo models and, more importantly, ~~can~~ could be applied to GCMs to improve our understanding of how BC in snow affects local hydrological cycles and the global climate.

Based on a comprehensive set of field measurements across the Northern Hemisphere, the BC coating effect in real snowpacks was evaluated by assuming the presence of an absorbing OC shell. The enhancement of snow albedo reduction ~~was~~ ranged from 1.13–1.27, and the enhancement of radiative forcing was 1.14–1.27, which exceeds the contribution of dust to snow light absorption over most areas of northern China. ~~Of note~~Notably, the greatest enhancements were detected on the Tibetan Plateau and in the Arctic, which ~~will~~ may likely contribute to further Arctic sea ice and Tibetan glacier retreat. Our findings indicated 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~~evaluate the climate on the Tibetan Plateau and Arctic more accurately.

## 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.

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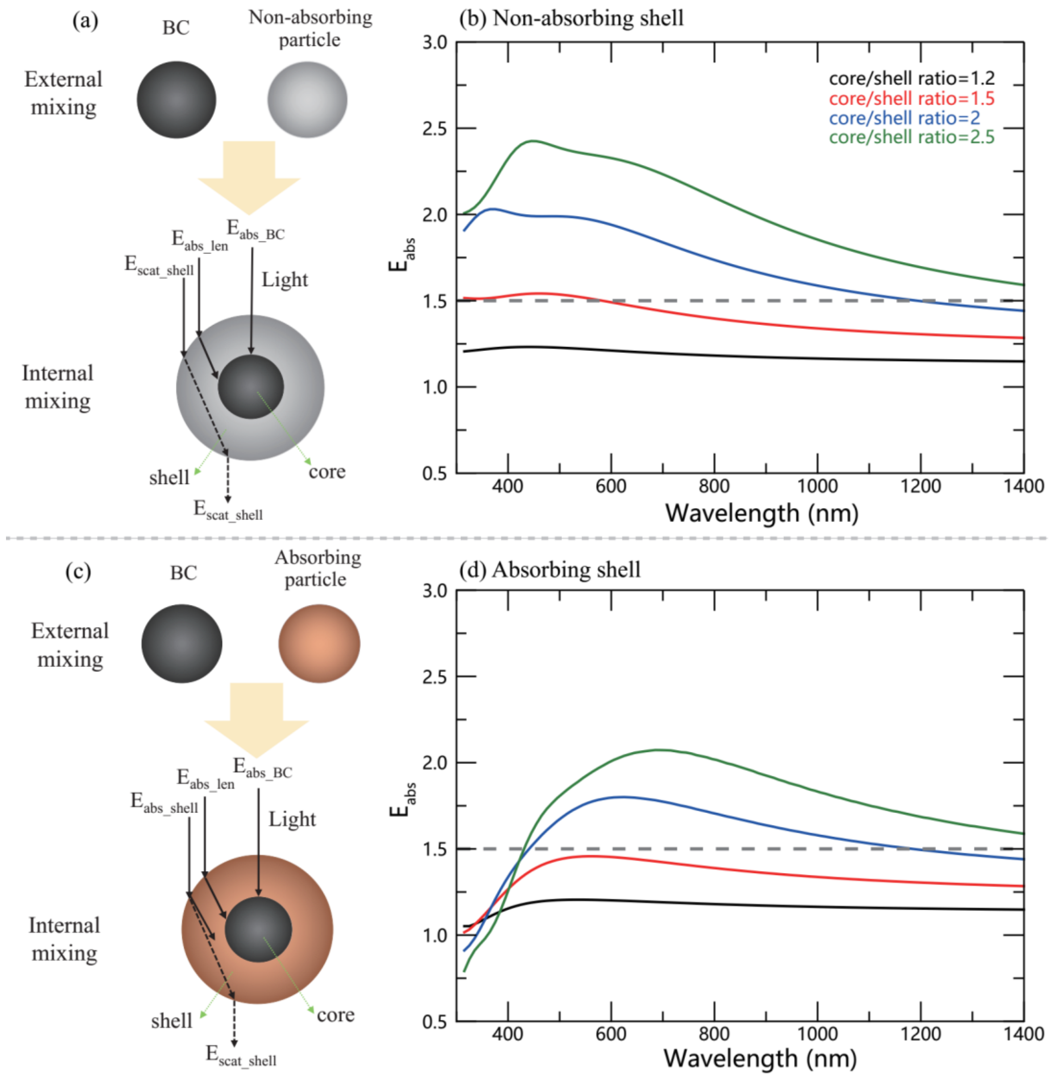

**Figure 1.** Schematic diagrams showing the light absorption ~~for~~ of an external mixture and internal mixture of BC~~,~~ ~~for~~ considering (a) a ~~non-absorbing~~nonabsorbing particle and (c) an absorbing particle. ~~Also shown is~~ Additionally, the enhancement of light absorption ~~from~~ due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

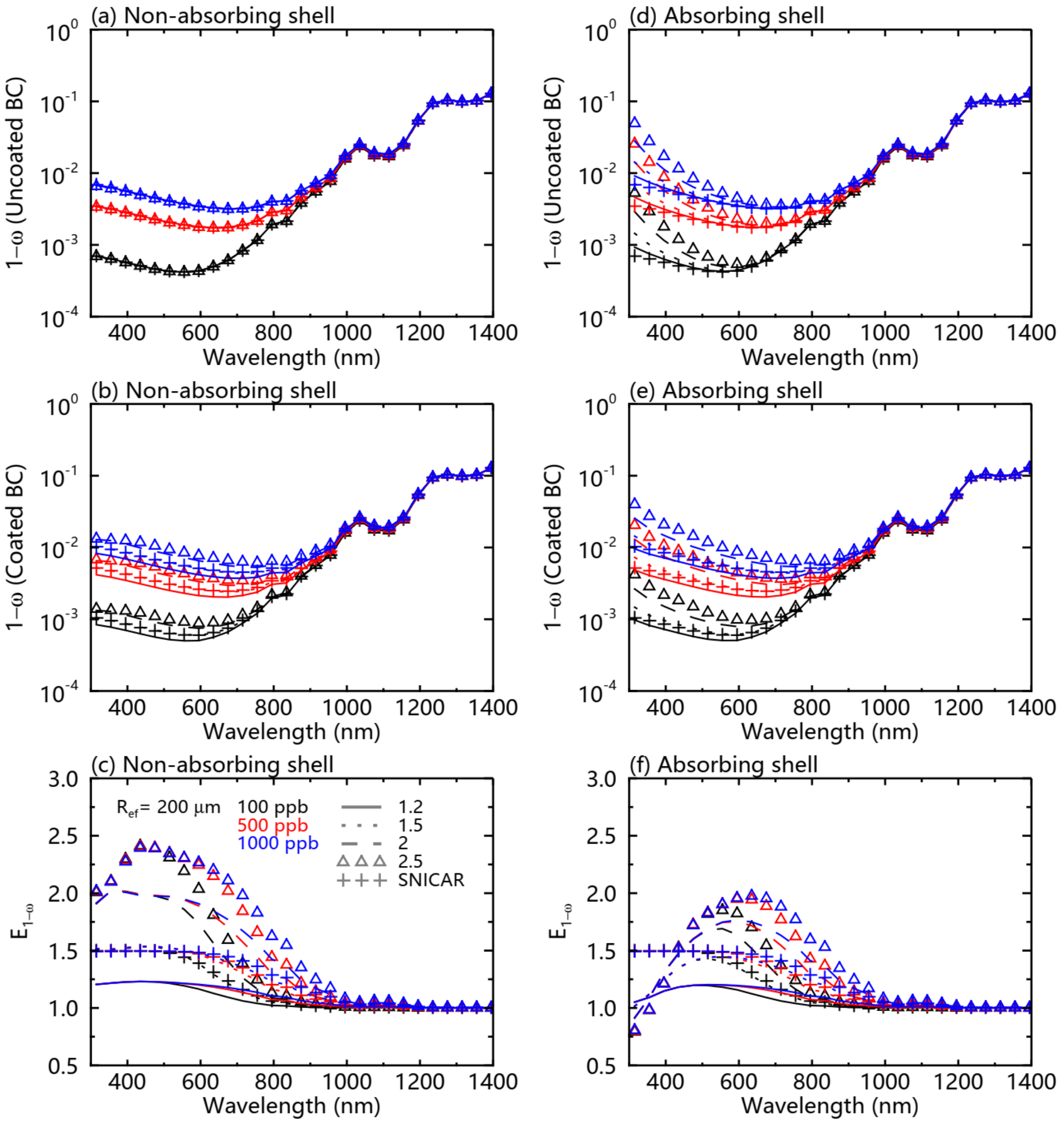

**Figure 2.** Snow single-scattering co-albedo (1–ω) as a function of the wavelength, ~~with~~ at different BC concentrations and core/shell ratios for (a) uncoated and (b) coated BC particles ~~with~~ under ~~an~~ the assumption of a ~~non-absorbing~~nonabsorbing shell. (d) and (e) are the same as (a) and (b), respectively, but ~~with~~ under ~~an~~ the assumption of an absorbing shell. (c) shows the ratios of the snow single-scattering co-albedo ($E_{1-\omega}$) for coated versus uncoated BC particles ~~with~~ under ~~an~~the assumption of a ~~non-absorbing~~nonabsorbing shell. (f) is the same as (c)~~,~~ but ~~with~~ under ~~an~~ the assumption of an absorbing shell. The snow grain radius was assumed to be 200 nm.

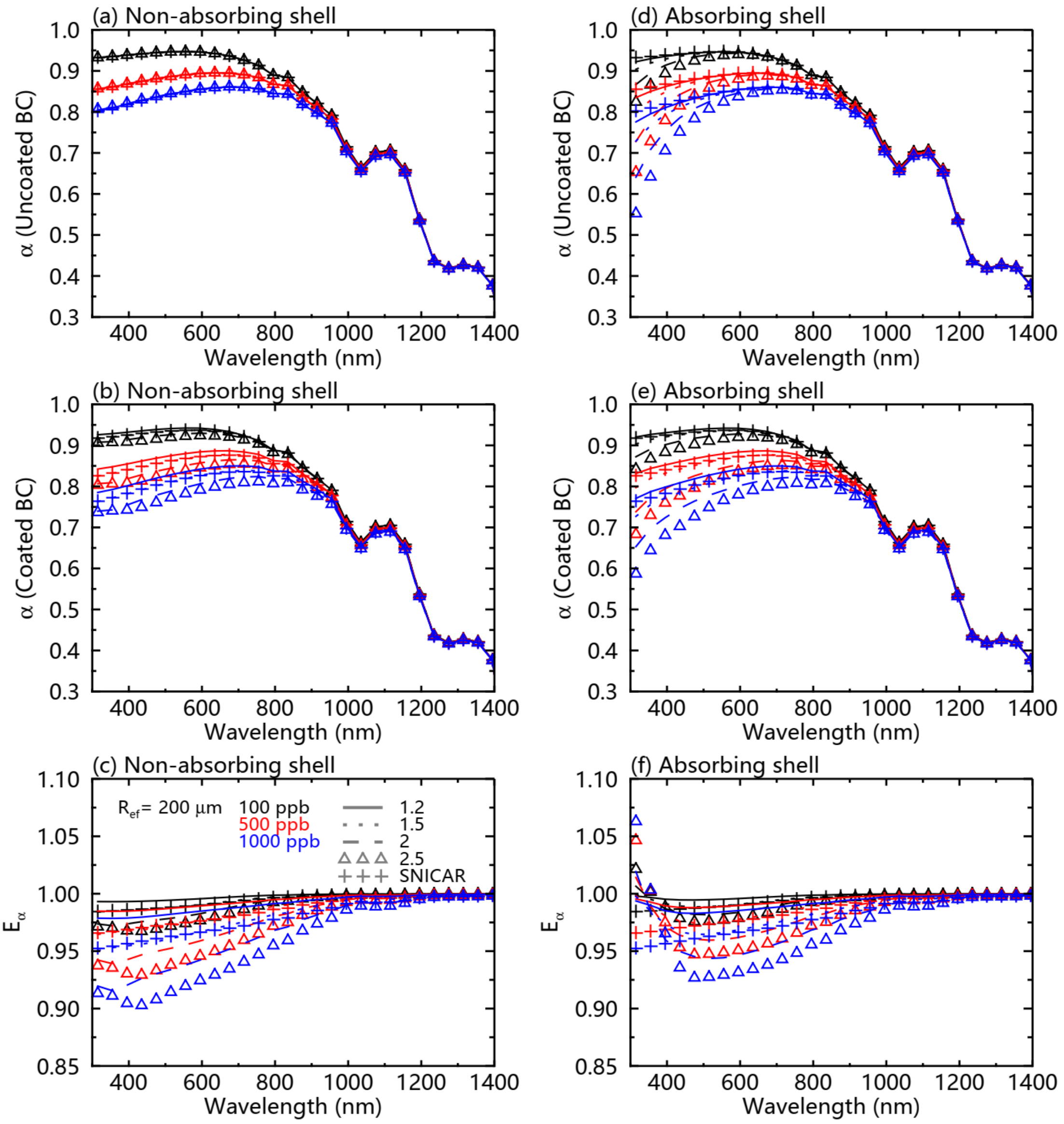

**Figure 3.** Same as Figure 2, but for the snow albedo (α).

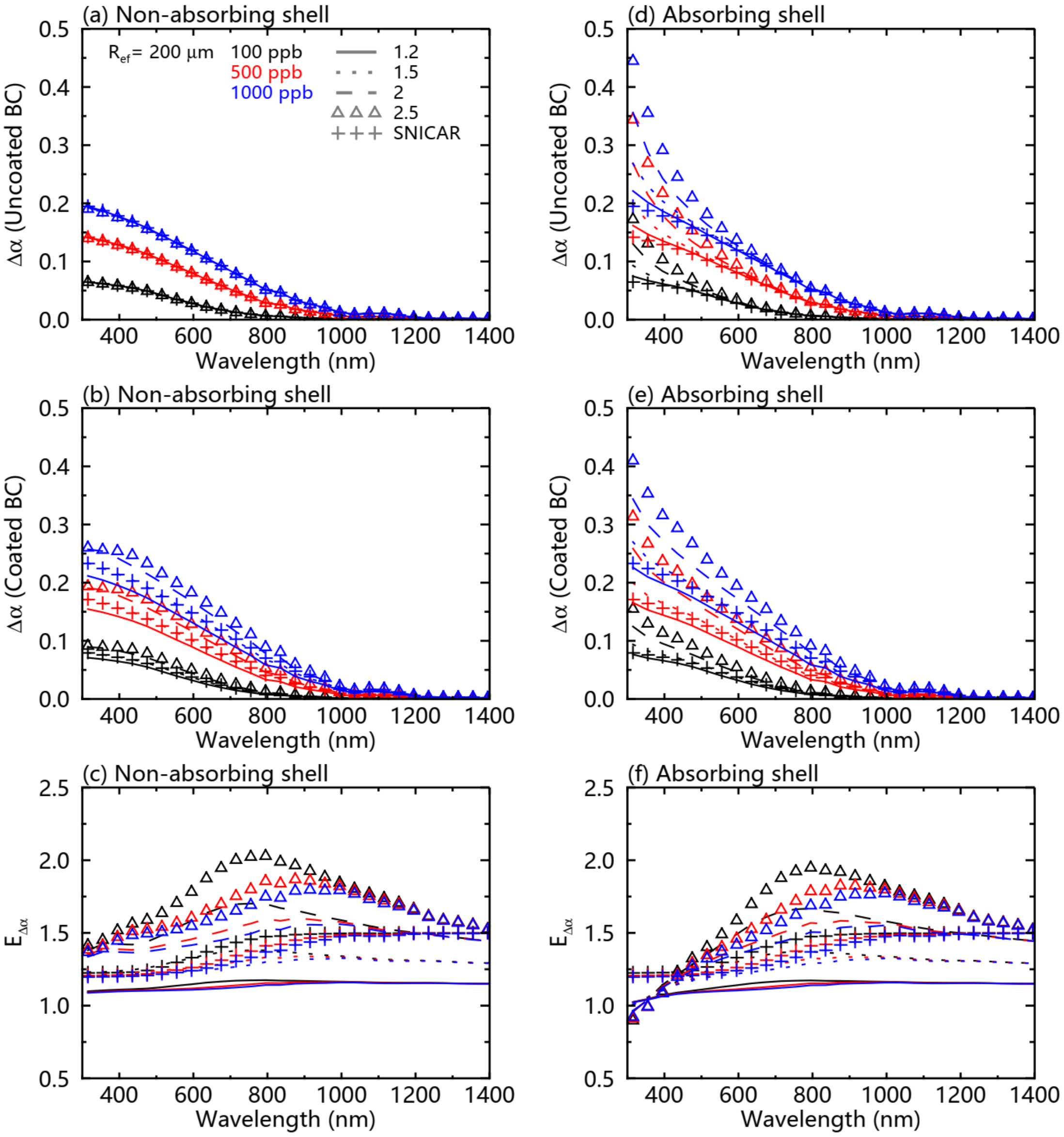

**Figure 4.** Same as Figure 2, but for the snow albedo reduction (Δα).

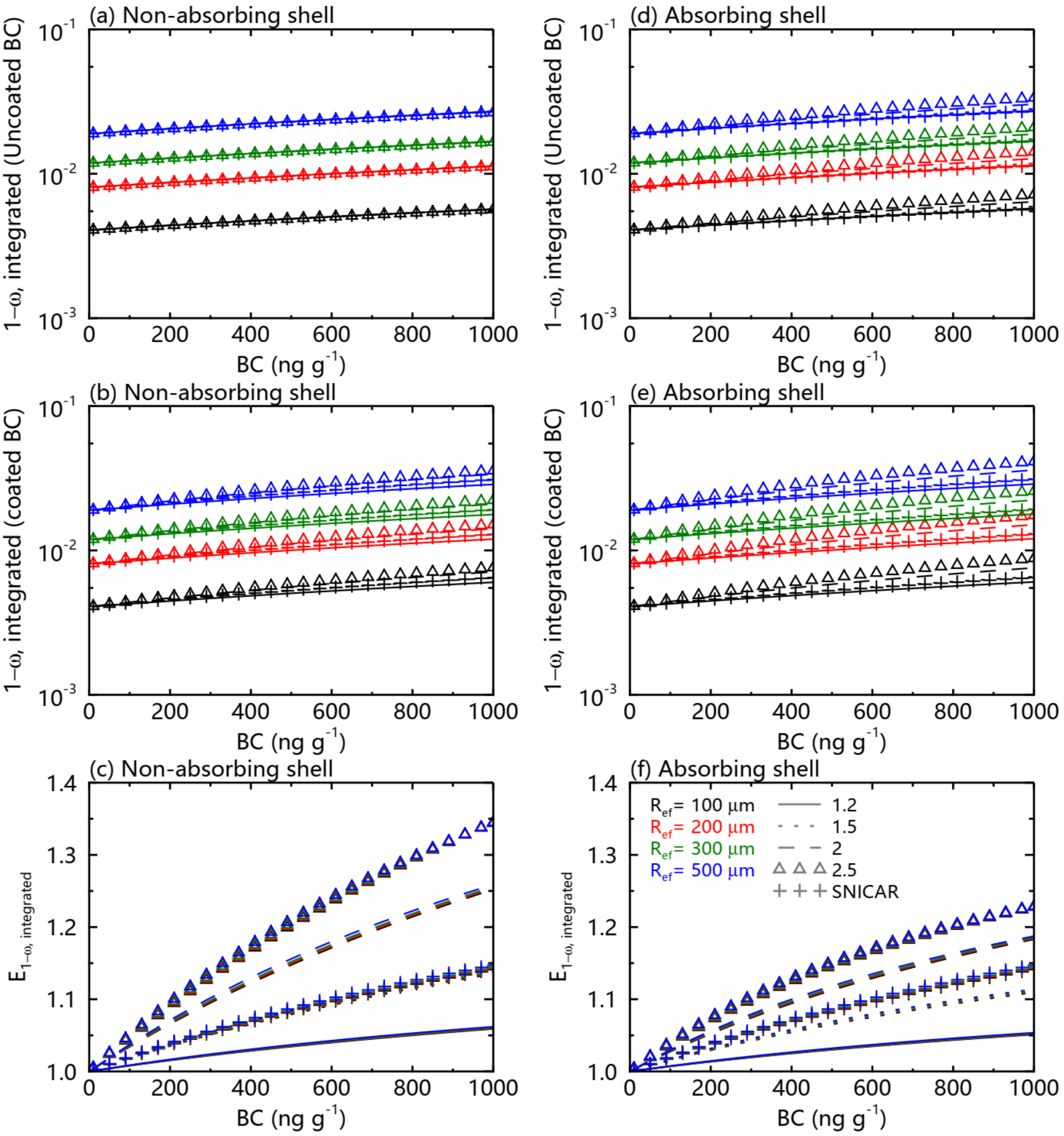

**Figure 5.** ~~The spectrally~~ Spectrally weighted snow single-scattering co-albedo (1-$\omega_{\text{integrated}}$) ~~over~~ from 300–2500 nm of ~~a~~ the typical surface solar spectrum at mid–high ~~latitude~~latitudes from January to May~~,~~ for (a) uncoated and (b) coated BC particles ~~with~~ under ~~an~~ the assumption of a ~~non-absorbing~~nonabsorbing shell. (d) and (e) are the same as (a) and (b), respectively, but ~~with~~ under ~~an~~ the assumption of an absorbing shell. (c) shows the ratios ($E_{1-\omega,\text{ integrated}}$) of the spectrally weighted snow single-scattering co-albedo for coated versus uncoated BC particles ~~with~~ under ~~an~~ the assumption of a ~~non-absorbing~~nonabsorbing shell. (f) is the same as (c)~~,~~ but ~~with~~ under ~~an~~ the assumption of an absorbing shell.

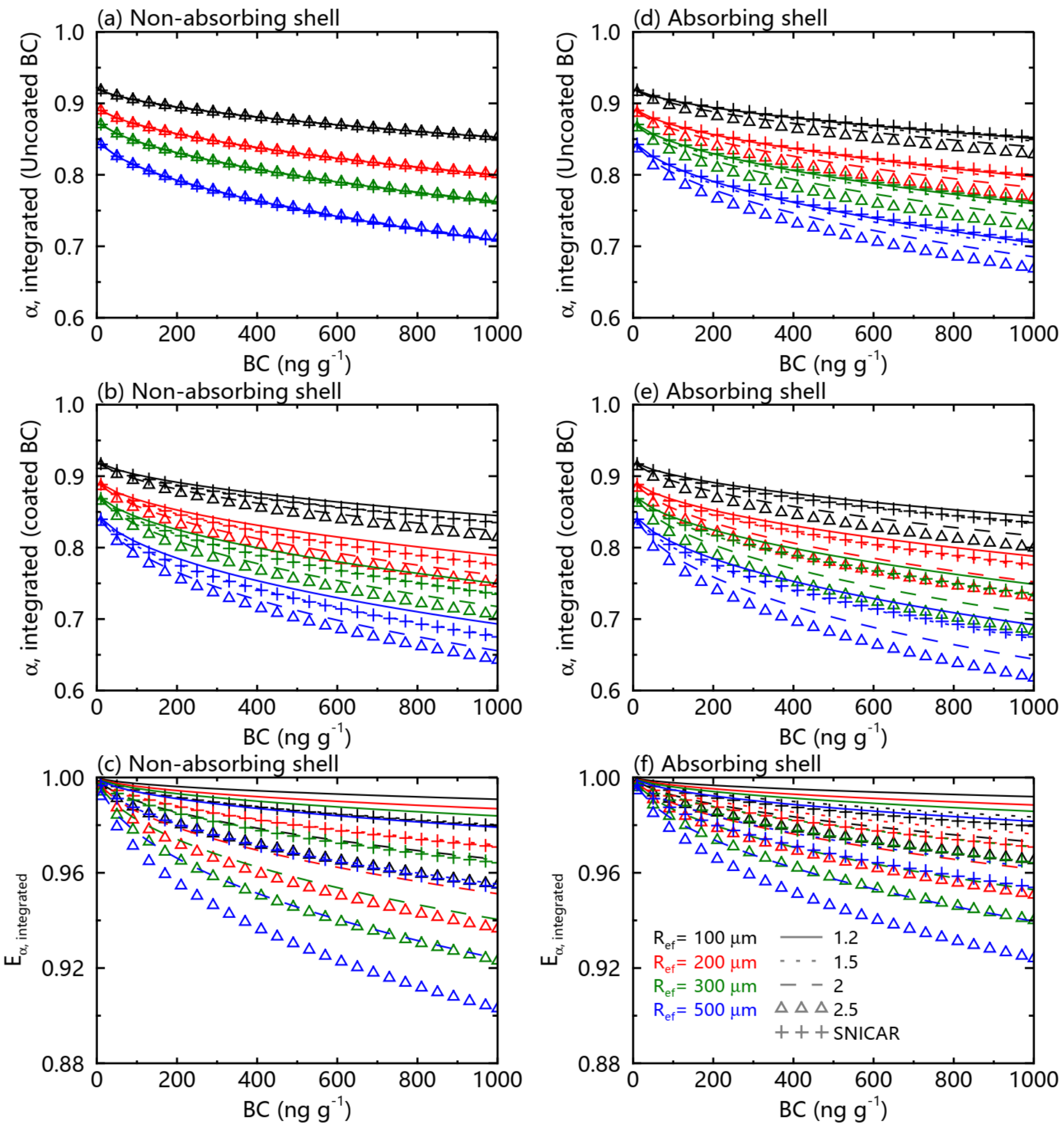

**Figure 6.** Same as Figure 5, but for the snow albedo ($\alpha_{\text{integrated}}$).

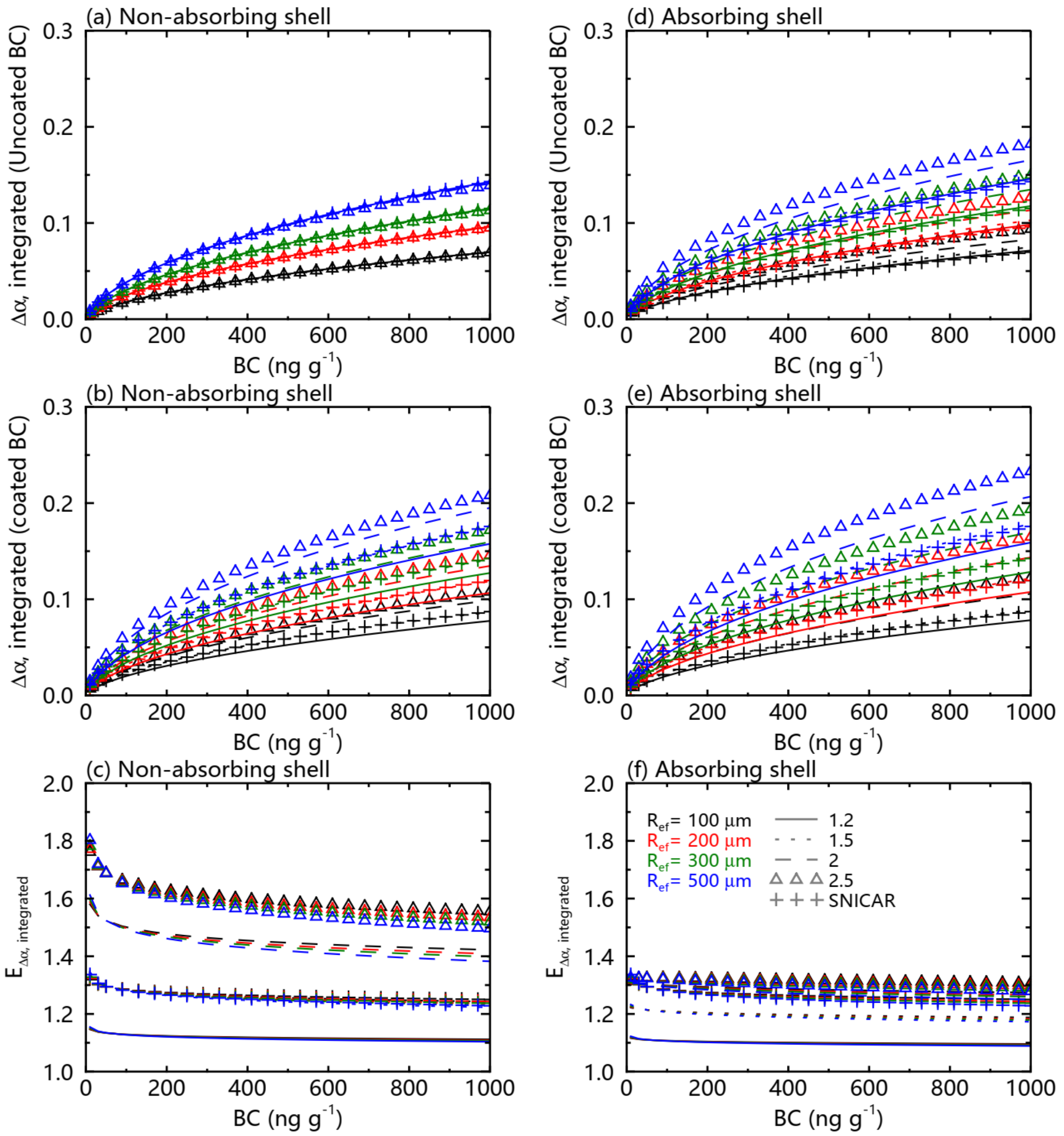

**Figure 7.** Same as Figure 5, but for the snow albedo reduction ($\Delta\alpha_{integrated}$).

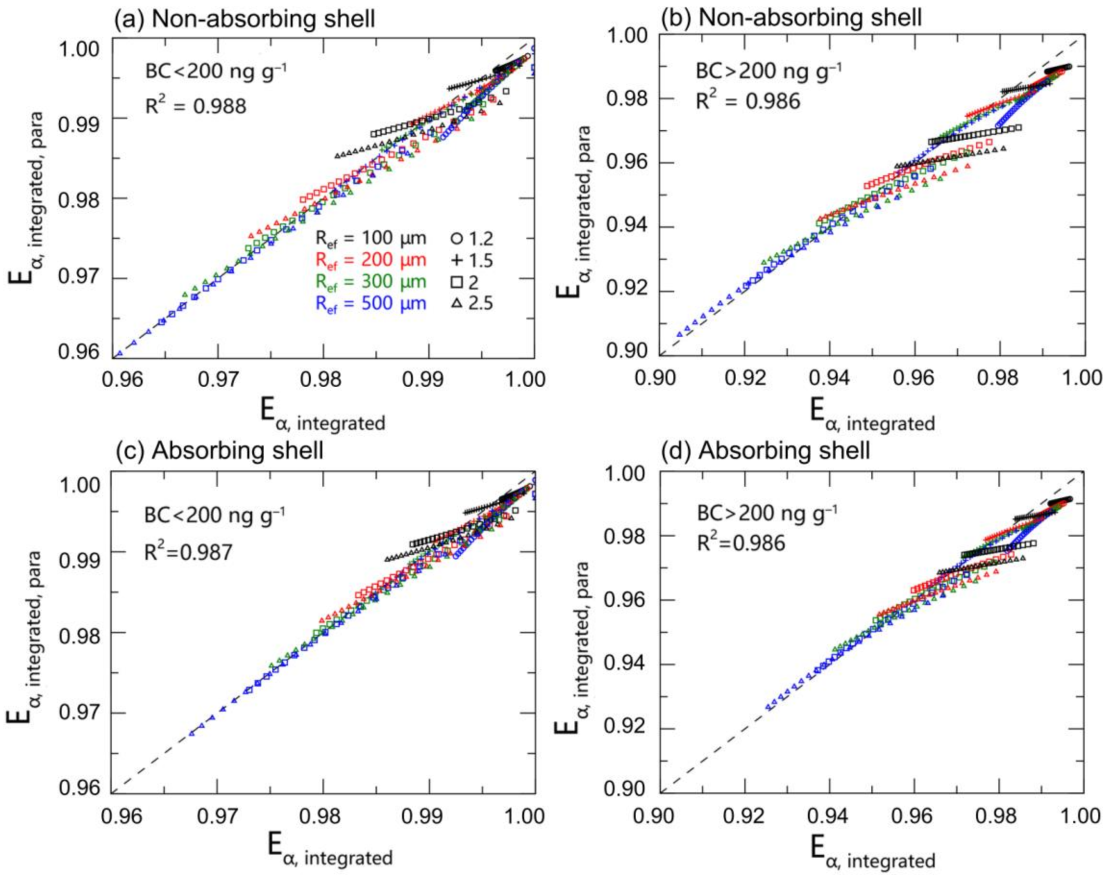

**Figure 8.** Comparisons of model~~-~~-calculated $E_{\alpha,\ \mathrm{integrated}}$ and parameterized $E_{\alpha,\ \mathrm{integrated,\ para}}$ values for (a) relatively clean snow (BC concentration <200 ng $g^{-1}$)~~,~~ and (b) relatively polluted snow (BC concentration >200 ng $g^{-1}$) for a ~~non-absorbing~~nonabsorbing shell. (c) and (d) ~~Same~~ are the same as (a) and (b), respectively, but for an absorbing shell.

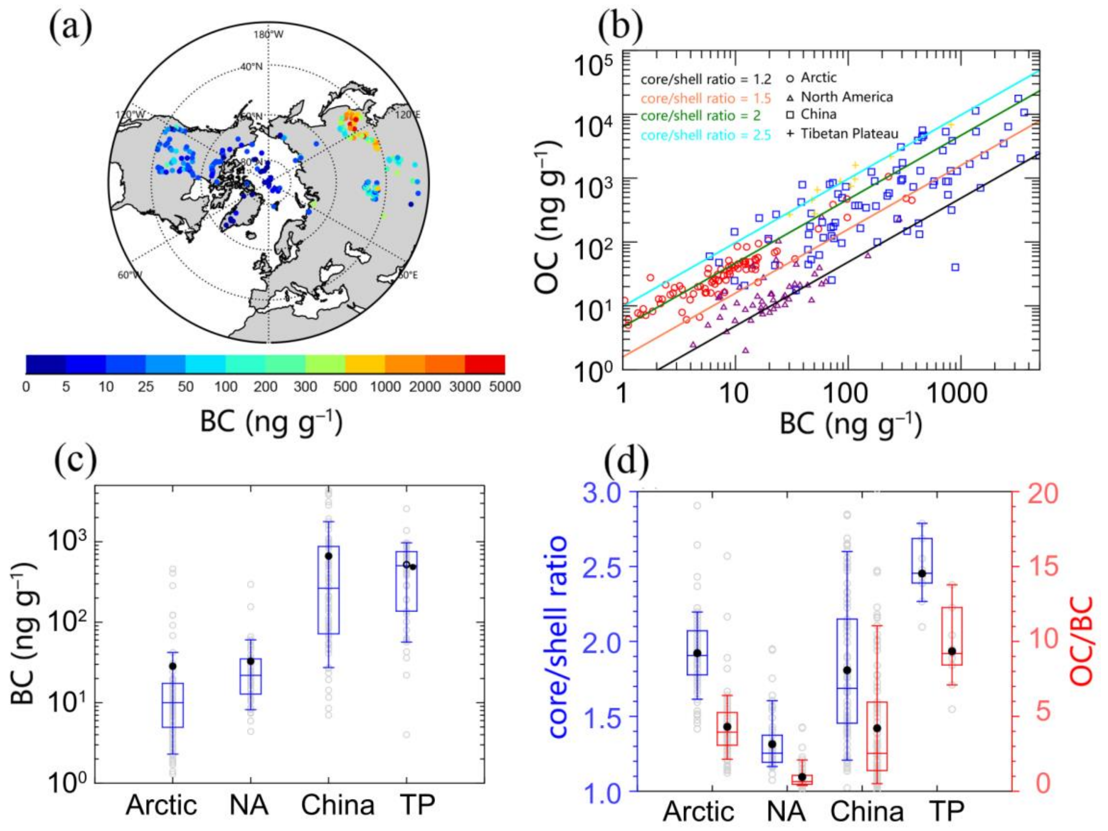

**Figure 9.** (a) ~~The s~~Spatial distribution of the measured black carbon (BC) concentration~~s~~ across the Northern Hemisphere. (b) Comparison of the BC and organic carbon (OC) concentrations in~~:~~ the Arctic, North America (NA), northern China (NC) and the Tibetan Plateau (TP). (c) Statistical plots of the BC concentration~~s~~ in the different regions. The boxes denote the 25th and 75th quantiles, the horizontal lines denote the 50th quantiles (median values), the solid dots denote the average values, and the whiskers denote the 10th and 90th quantiles. ~~In~~ The in situ data ~~is~~ are shown as gray circles. (d) ~~Same~~ is the same as (c) but for ~~a~~ the core/shell ratio and OC/BC mass ratio, assuming a core/shell structure with a BC core and an absorbing OC shell.

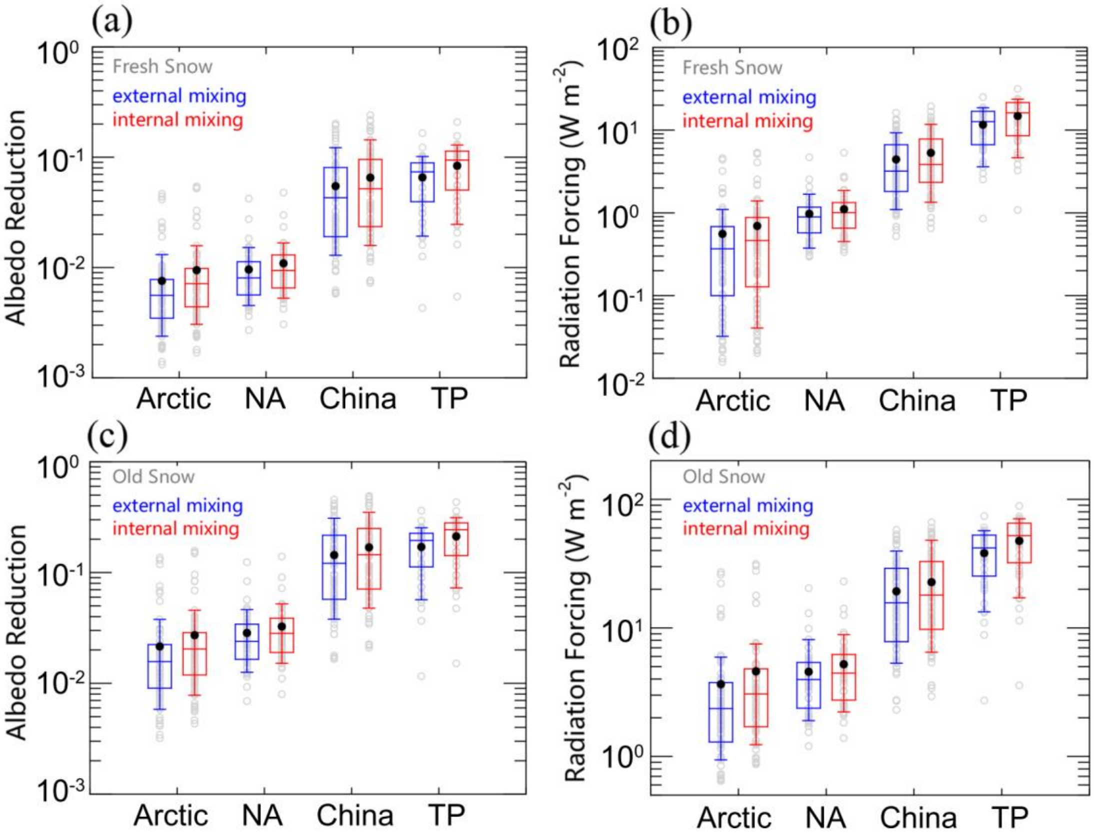

**Figure 10.** Statistical plots of (a) albedo reduction~~,~~ and (b) radiative forcing~~,~~ in the different regions for fresh snow. (c) and (d) ~~Same~~ are the same as (a) and (b), respectively, but for old snow. The boxes denote the 25th and 75th quantiles, the horizontal lines denote the 50th quantiles (median values), the solid dots denote the average values, and the whiskers denote the 10th and 90th quantiles. ~~In~~ The in situ data ~~is~~ are shown as gray circles.

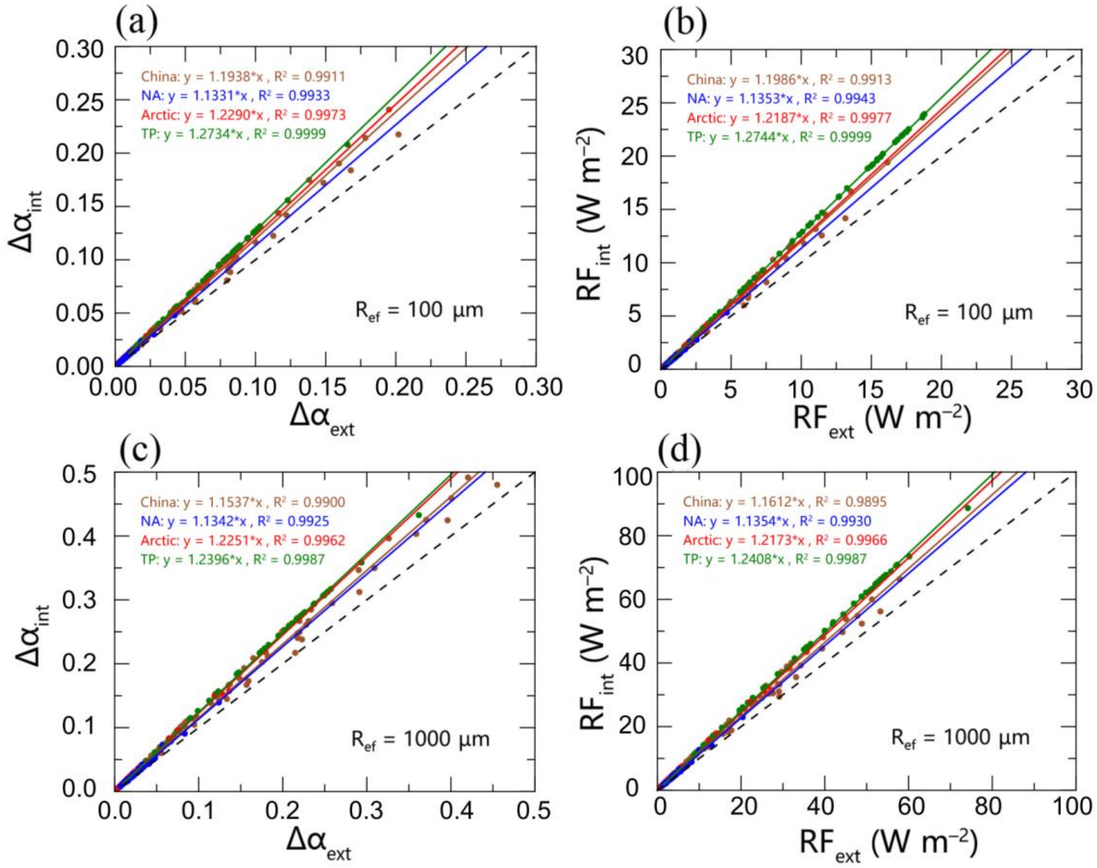

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**Figure 11.** Comparisons of (a) ~~the~~ snow albedo reduction and (b) ~~the~~ radiative forcing ~~by an~~via internally mixed particles versus ~~an~~ external mixed particles~~,~~ based on in situ measurements of fresh snow (assuming a snow grain radius of 100 μm). (c) and (d) ~~Same~~ are the same as (a) and (b), respectively,~~,~~ but for old snow and assuming a snow grain radius of 1000 μm.