

## Response to Cenlin He's comments

We thank Cenlin He for his comments, which have helped us improve the paper quality. We have incorporated these comments into the revised paper as detailed below.

The authors conducted extensive field measurements of BC, OC, and dust concentrations in snow over northern China and employed the SNICAR and SAMDS model simulations to investigate snow albedo reduction caused by light-absorbing aerosols. This study provides a valuable observational dataset to improve our understanding of the effects of light-absorbing aerosol deposition on snow albedo reduction. I have a short comment on snow albedo modeling. The authors mentioned that the SAMDS model considers aerosol-snow mixing state and the irregular morphology of snow grain by using asymptotic radiative transfer theory. However, the authors did not provide enough discussions on the effects of BC/dust/OC-snow mixing state and snow grain shape on snow albedo reduction as well as compare with some recent studies. These factors are not trivial in evaluating aerosol-snow albedo effects, in addition to snow grain size and aerosol concentration. For example, recent studies by Liou et al. (2014) and He et al. (2014) developed and applied a stochastic snow model to study BC/dust-induced snow albedo reduction, which explicitly simulates different aerosol-snow mixing states and snow grain shapes. They found that using a realistic snowflake shape reduces BC-induced snow albedo reduction by 20–40% compared to a spherical snow grain, while multiple internal mixing of BC and snow increases the albedo reduction by 40–60% relative to the external mixing. I would suggest discussing these recent findings, which could be very helpful for people to understand potential uncertainty and improvement in snow albedo modeling.

R: We noted that the two references by Liou et al. (2014) and He et al. (2014) are both very important achievements for understanding light absorption by internally mixed BC/dust in snow grains for application to climate models. Therefore, we also used the SAMDS model to simulate the snow albedo change by internal/external mixing of BC and snow associated with the irregular morphology of snow grains using asymptotic radiative transfer theory (Figure 10). The following discussions on snow albedo change due to internal/external mixing states of ILAPs associated with irregular snow grains in snow have been added in the revised manuscript.

### 1. Introduction

“Warren and Wiscombe (1980) found that a mixing ratio of  $10 \text{ ng g}^{-1}$  of soot in snow can reduce snow albedo levels by 1%. Light et al. (1998) determined that  $150 \text{ ng g}^{-1}$  of BC embedded in sea ice can reduce ice albedo levels by a maximum of 30%.  $1 \text{ ng g}^{-1}$  of BC has approximately the same effect on the albedo of snow and ice at 500 nm as  $50 \text{ ng g}^{-1}$  of dust (Warren, 1982). Doherty et al. (2013) analyzed field measurements of vertical distributions of BC and other ILAPs in snow in the Arctic during the melt season and found significant melt amplification due to an increased

mixing ratio of BC by up to a factor of 5. Yasunari et al. (2015) suggested that the existence of snow darkening effect in the Earth system associated with ILAPs contributes significantly to enhanced surface warming over continents in northern hemisphere midlatitudes during boreal spring, raising the surface skin temperature by approximately 3–6 K near the snowline. Warren and Wiscombe (1985) pointed out that modeling soot in snow as an “external mixture” (impurities particles separated from ice particles) may underestimate the true effect of the impurities as a given reduction of albedo by about half as much soot, if the soot is instead located inside the ice grains as an “internal mixture”. Hansen et al. (2004) and Cappa et al. (2012) noted that for a given BC mass on snow albedo, the internal mixing of BC in snow is a better approximation than external mixing, whereas internal mixing increases the BC absorption coefficient by a factor of two, for better agreement with empirical data. Hadley and Kirchstetter (2012) also indicated that increasing the size of snow grains could decrease snow albedo and amplify radiative perturbation of BC. For a snow grain optical effective radius ( $R_{\text{eff}}$ ) of 100  $\mu\text{m}$ , the albedo reduction caused by 100  $\text{ng g}^{-1}$  of BC is 0.019 for spherical snow grains but only 0.012 for equidimensional nonspherical snow grains (Dang et al., 2016). Fierce et al. (2016) pointed out that BC coated with non-absorbing particles absorbs more strongly than the same amount of BC in an uncoated particle, but the magnitude of this absorption enhancement is still a challenge. He et al. (2014) indicated that BC-snow internal mixing increases the albedo forcing by 40–60% compared with external mixing, and coated BC increases the forcing by 30–50% compared with uncoated BC aggregates, whereas Koch snowflakes reduce the forcing by 20–40% relative to spherical snow grains using a global chemical transport model in conjunction with a stochastic snow model and a radiative transfer model.”

## 2.5 Model simulations

The contents of the asymmetry factor for calculating the snow albedo change by irregular snow grains have been added in Section 2.5 as: “The theory based on the ray-optics approach shows that  $g$  in Equation (11) and  $B$  in Equation (13) are 0.89 and 1.27 for spheres, 0.84 and 1.50 for hexagonal plates/columns, and 0.75 and 1.84 for fractal grains, respectively.”

## 3.3 Simulations of snow albedo

“Previous studies indicated that the mixing ratio of BC (10-100  $\text{ng g}^{-1}$ ) in snow may decrease its albedo by 1-5% (Hadley and Kirchstetter, 2012; Warren and Wiscombe, 1980). Liou et al. (2011) demonstrated that a small BC particle on the order of 1  $\mu\text{m}$  internally mixed with snow grains could effectively reduce visible snow albedo by as much as 5-10% using a geometric-optics surface-wave approach. They also found that internal mixing of BC in snow reduces snow albedo substantially more than external mixing, and the snow grain shape plays a critical role in snow albedo calculations through its forward scattering strength by modeling the positions of BC internally mixed with different snow grain types (Liou et al., 2014).

Figure 10a illustrates the effect of snow shape (fractal grains, hexagonal

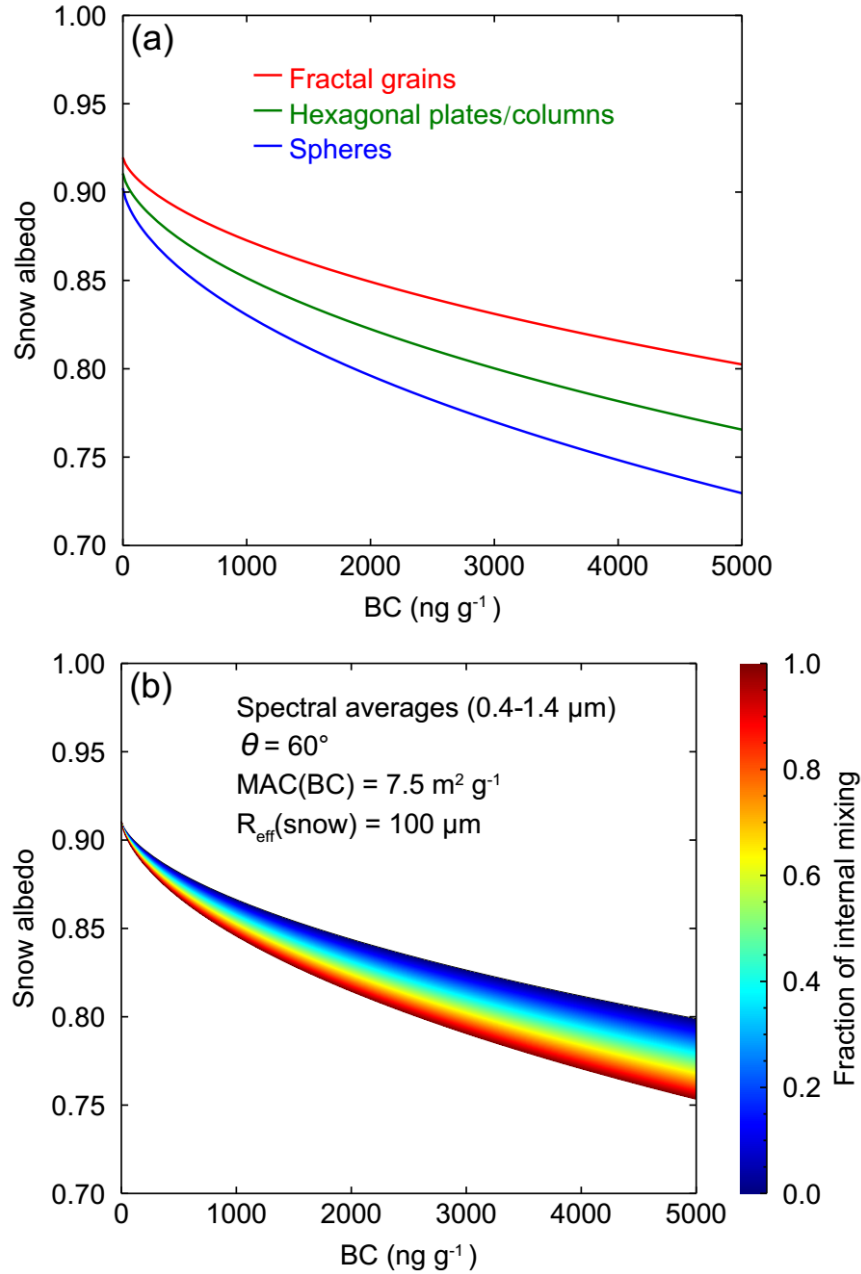
plates/columns, and spheres) on snow albedo at the spectral wavelengths of 400 nm-1400 nm with  $R_{\text{eff}}$  of 100  $\mu\text{m}$  simulated by SAMDS model. As is shown, the differences of snow albedo caused by three snow shapes are remarkable. The snow albedo for spherical snow grains is higher than that for the other two shapes because the scattering by spherical snow grains is more in forward direction and less to the sides, resulting in a larger  $g$  and a smaller  $B$  as discussed in section 2.5. In addition, the snow albedo reduction for aged snow such as spherical snow grains is larger than fresh snow such as fractal snow grains, and hexagonal plates/columns snow grains with the increased BC in snow. It shows that snow albedo by spherical snow grains is typically lower by 0.017-0.073 than the fractal snow grains, and by 0.008-0.036 than the hexagonal plates/columns snow grains, as a function of BC mixing ratios (0-5000  $\text{ng g}^{-1}$ ). Dang et al. (2016) assessed the effects of snow grain shapes on snow albedo using the asymmetry factors  $g$  of nonspherical ice crystal developed by Fu (2007). They obtained similar result that the albedo reduction caused by 100  $\text{ng g}^{-1}$  of BC for spherical snow grains is larger by 0.007 than nonspherical snow grains with the same area-to-mass ratio for  $R_{\text{eff}}$  of 100  $\mu\text{m}$ .

Figure 10b shows the spectral albedo of snow for the internal/external mixing of BC and snow with  $R_{\text{eff}}$  of 100  $\mu\text{m}$  for a solar zenith angle  $\theta$  of  $60^\circ$  as a function of BC mixing ratio. For a given shape (hexagonal plates/columns), we found that snow albedo as a function of BC mixing ratios calculated from this study decreases as the fraction of the internal mixing increases (Figure 10b). In previous studies, the BC mixing ratios in seasonal snow were up to 3000  $\text{ng g}^{-1}$  due to heavy industrial activities across northern China, but the lowest mixing ratios of BC were found in the remote northeastern on the border of Siberia, with a median value in surface snow of 100  $\text{ng g}^{-1}$  (Huang et al., 2011; Wang et al., 2013a, 2014; Ye et al., 2012). As a result, snow albedo by internal mixing of BC and snow is lower than external mixing by up to 0.036 for 3000  $\text{ng g}^{-1}$  BC in snow in the heavy industrial regions across northeastern China, whereas by low to 0.005 for 100  $\text{ng g}^{-1}$  BC in snow in the further north China near the border of Siberia. We indicated that the snow grain shape effect on snow albedo between spherical snow grains and fractal snow grains is relatively larger than the effect of the internal/external mixing of BC and snow as a function of the BC mixing ratios. However, He et al. (2014) also pointed out that the snow albedo reductions computed by previous models under assorted assumptions vary by a factor of 2 to 5.”.

#### 4 Conclusions

“Then, we indicated that the new spectral snow albedo model (SAMDS) can be used to investigate the snow albedo influenced by the internal/external mixing of BC and snow, irregular morphology of snow grains and the vertical distribution of snow grains. For instance, the snow albedo for spherical snow grains is typically lower than that for the fractal snow grains and hexagonal plates/columns snow grains with  $R_{\text{eff}}$  of 100  $\mu\text{m}$ . The internal mixing of BC and snow absorbs substantially more light than external mixing. For fresh snow grains of hexagonal plates/columns with  $R_{\text{eff}}$  of 100  $\mu\text{m}$ , the difference of snow albedo between internal and external mixing of BC and

snow is up to 0.036 for 3000 ng g<sup>-1</sup> BC in snow in the heavy industrial regions across northeastern China, whereas by low to 0.005 for 100 ng g<sup>-1</sup> BC in snow in the further north China near the border of Siberia. The spectral albedo of snow reduction caused by OC (20 µg g<sup>-1</sup>) is larger by up to a factor of 3 for a snow grain size of 800 µm compared to 100 µm by using SAMDS model.”



**Fig. 10.** Spectral albedo of snow as a function of BC mixing ratios in snow by using SMDAS model for: (a) the irregular morphology of snow grains (fractal grains, hexagonal plates/columns, and spheres), (b) internal/external mixing of BC and hexagonal plates/columns snow grains. Also shown are model parameters including integrate spectral wavelengths (400-1400 nm), solar zenith angle ( $\theta$ ), mass absorption coefficient (MAC) of BC, and snow grain optical effective radius ( $R_{\text{eff}}$ ).

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