Anonymous Referee #4

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General comments

This paper reports a comparison of the impacts on snow albedo due to dust externally vs. internally mixed with snow, starting from idealized cases and then proceeding to estimates of albedo reduction based on actual measurements of dust concentration in snow at various geographical locations in the Northern hemisphere.

This topic is relevant for ACP and the paper is mostly well-written, but unfortunately there is one major concern (as also noted by the first reviewer). The Maxwell-Garnett approximation is applied to cases which are, in principle, much outside its region of validity. Therefore, I can recommend the publication of this paper only if the authors are able to use a more rigorous approach, or — at very minimum — to carefully evaluate the accuracy of their results by comparison to more rigorous calculations found in the literature.

R: We agreed with the reviewer that the Maxwell-Garnett approximation has been well applied in analyzing the BC/snow internal mixing. However, previous studies also acknowledged that the Maxwell-Garnett approximation can create some errors for the albedo calculation of dust/snow internal mixing (Flanner et al., 2012). Therefore, we have carefully evaluated the accuracy of our results based on the effective medium approximation by comparison to more rigorous calculations found in He et al. (2019), which used the geometric-optics surface-wave approach (GOS) to consider the impact of dust/snow internal mixing on snow absorption and albedo reduction. As shown in Figure R1 (i.e., Figure 11 in the revised manuscript), the enhancement ratio of snow albedo reduction (1.28) due to dust–snow internal mixing (relative to external mixing) in this study is slightly higher than the value (1.16) reported by He et al. (2019), and this deviation is comparable to that caused by snow nonsphericity (He et al., 2019). Therefore, we indicated that the effective medium approximation used in this study is reasonable and reliable. The detailed description can be found in p. 19, lines 1-12.

"In this study, the application of the effective medium approximation greatly simplifies

the complexity of snow radiation transfer calculation for dust–snow internal mixing, and the effect of non-uniform distribution of dust in snow grains on snowpack optical properties are explicitly quantified for the first time. However, it is worth noting that this method has its limitation when applying to large particles (e.g., dust) in snow (Bohren 1986; Flanner et al., 2012), which can create some errors for the albedo calculation of dust–snow internal mixing. To verify the credibility of our results, we carefully make a comparison with the more rigorous calculations found in He et al. (2019), which used the geometric-optics surface-wave approach (GOS) to consider the impact of dust–snow uniform internal mixing on snow albedo reduction. As shown in Figure 11, the results show that the enhancement ratio of snow albedo reduction (1.28) due to dust–snow internal mixing (relative to external mixing) is slightly higher than the value (1.16) reported by He et al. (2019), and this deviation is comparable to that caused by snow nonsphericity (He et al., 2019). Therefore, we indicated that the effective medium approximation used in this study is reasonable and reliable.”

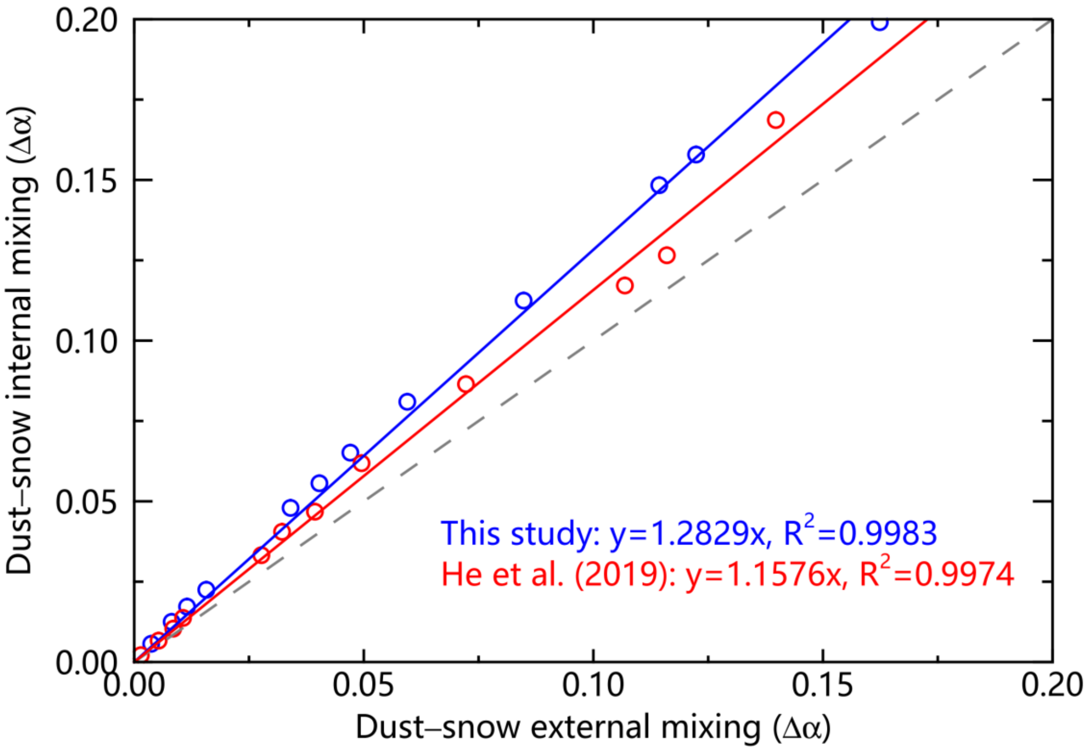

Figure R1. Comparisons of snow albedo reduction ($\Delta\alpha$) under the cloudy sky caused by dust-snow uniform internal mixing (y-axis) and external mixing (x-axis) for this study (blue) and He et al. (2019) (red).

Major comments

1. The use of the Maxwell-Garnett approximation to compute the effective refractive index (and subsequently the single-scattering properties) of snow grains containing mineral dust particles is physically questionable. The problem is that the Maxwell-Garnett approximation assumes that the inclusions are much smaller than the wavelength. In the case of dust particles, with an effective radius of 1.1 μm as considered here, this is definitely not the case (e.g., the effective size parameter $x = 2\pi r/\lambda$ exceeds 10 in the visible region).

Consequently, when comparing the effects of dust internally vs. externally mixed with snow, you are in fact comparing dust particles with different size: particles in the micrometer scale for external mixing, and (in principle) infinitesimally small particles for internal mixing.

R: To better explore the effects of different dust size on snow albedo, we provide a case study of albedo calculation for dust-snow external/internal mixing with an effective dust radius of 0.1 μm (similar to the size of black carbon) shown in Figure R2. Owing to the dust size used in this case is really small, we presumed that the comparison between dust-snow external mixing and internal mixing is performed under the same dust size, and the difference can be regarded as a reference standard. Meanwhile, the results of snow albedo for other three dust sizes (1.1, 2.5, and 5.0 μm) were also presented in Figure R2, we noted that the differences induced by different dust sizes were far less than those induced by the dust-snow mixing state. Therefore, we indicated that the effects of dust size on the comparison between dust-snow external mixing and internal mixing were limited, and the results of this study were acceptable.

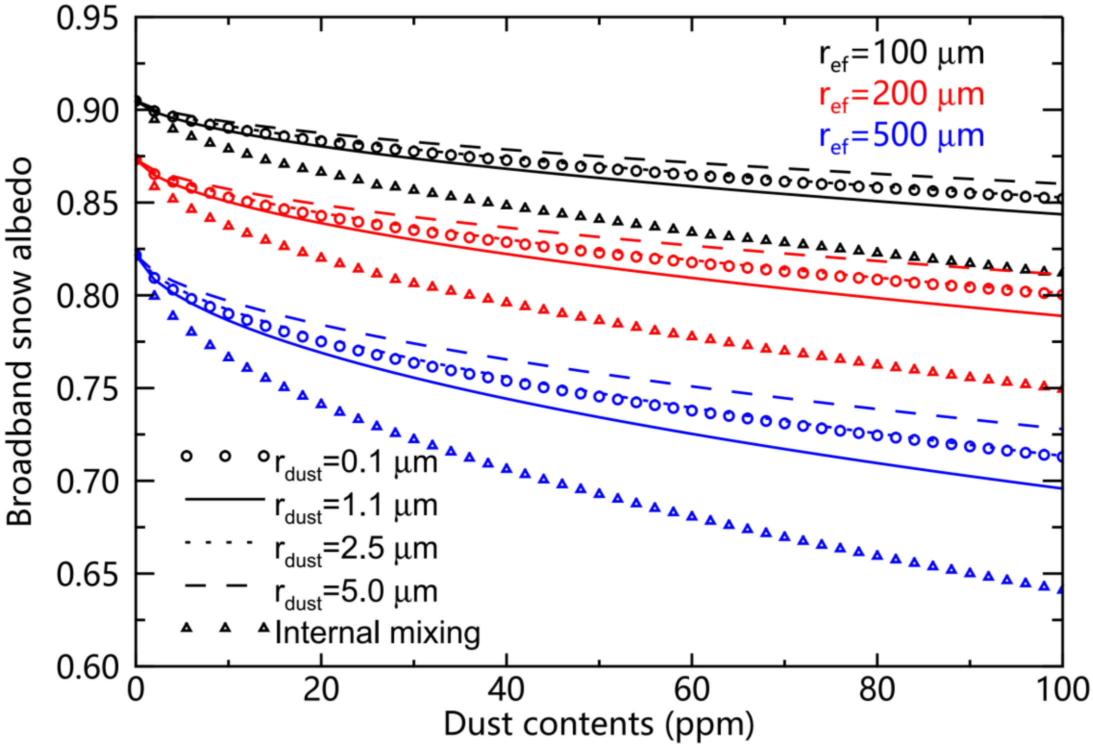

Figure R2. The broadband snow albedo variations with dust contents due to four different dust sizes (0.1, 1.1, 2.5, and 5.0 μm) for dust-snow external mixing, and the results for dust-snow internal mixing were also presented.

2. Neither the introduction nor other parts of the paper discuss the present work properly in the context of previous studies that have considered internal mixing of dust within snow grains. These include, at least, the studies by Liou et al. (2014) and He et al. (2019), both of which appear in the reference list of the current paper (so the authors seem to be aware of their existence anyway).

These papers use a more rigorous approach (the geometric-optics surface-wave approach, GOS) and they also consider the impact of snow grain shape but not whether the dust particles are concentrated in the inner or outer parts of snow grains. Also note that the paper by He et al. (2019) employs the same size distribution for dust as the current paper. This should allow a comparison of the results of your approach with the GOS approach.

R: We have added more contents to analyze the related studies about dust-snow internal mixing effects from Liou et al. (2014) and He et al. (2019), which can be found in p. 4, lines 23-28:

"Liou et al. (2014) is the pioneer in investigating dust-snow internal mixing effects based on the geometric-optics surface-wave approach. Subsequently, He et al. (2019) used the same method to explicitly quantify the combined effects of dust-snow internal mixing and snow grain nonsphericity on snow optical properties, thereafter, develop a set of new dust-snow parameterizations for land/climate modeling applications for the first time."

Besides, we also carefully evaluated the accuracy of our results by comparison to more rigorous calculations found in He et al. (2019). The detailed description can be found in p. 19, lines 1-12:

"In this study, the application of the effective medium approximation greatly simplifies the complexity of snow radiation transfer calculation for dust–snow internal mixing, and the effect of non-uniform distribution of dust in snow grains on snowpack optical properties are explicitly quantified for the first time. However, it is worth noting that this method has its limitation when applying to large particles (e.g., dust) in snow (Bohren 1986; Flanner et al., 2012), which can create some errors for the albedo calculation of dust–snow internal mixing. To verify the credibility of our results, we carefully make a comparison with the more rigorous calculations found in He et al. (2019), which used the geometric-optics surface-wave approach (GOS) to consider the impact of dust–snow uniform internal mixing on snow albedo reduction. As shown in Figure 11, the results show that the enhancement ratio of snow albedo reduction (1.28) due to dust–snow internal mixing (relative to external mixing) is slightly higher than the value (1.16) reported by He et al. (2019), and this deviation is comparable to that caused by snow nonsphericity (He et al., 2019). Therefore, we indicated that the effective medium approximation used in this study is reasonable and reliable."

Minor comments

1. p. 5, line 27: There is an error in Eq. (3). It should be $\sigma_{ext} = \frac{1}{l_{tr}(1-g)} = \frac{3C_v}{2r_{ef}}$. See Eq. (18) in Kokhanovsky and Zege (2004). This is also obvious if you consider the units.

R: Changed as suggested.

2. p. 9, lines 2–5: Was the effect of clouds included in the calculation of the solar spectrum? The reason why I'm asking is that the broadband albedo for pure snow in Fig. 10 (about 0.87) seems quite high (i.e., too high for cloud-free conditions, for $r_{ef} = 200\ \mu m$).

R: The incoming solar irradiance used in this study is a typical surface solar spectrum at mid to high latitudes from January to March under the cloudy sky, calculated by the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model. This related description has been added in the revised manuscript (See Page 9, Line 9-12).

3. p. 10, line 17: "the order of magnitude of k_{ice} was comparable to k_{dust} at those wavelengths". This is not quite true, as the difference between k_{dust} and k_{ice} is still 2–3 orders of magnitude. It is just more than compensated by the much larger difference in ice vs. dust concentration.

R: The sentence has been rewritten in p. 10, lines 22-26:

"Additionally, it is worth noting that k_{ef} was not sensitive to dust mass concentrations in wavelengths >1000 nm, which was generally consistent with k_{ice}, because the difference between k_{dust} and k_{ice} is more than compensated by the much larger difference in ice and dust concentration at those wavelengths."

4. p. 12, line 25: "monotonic dependence of σ_{abs}" on what? On $\overline{r_c}$ or $\overline{r_p}$?

R: Changed as suggested.

"… monotonic dependence of σ_{abs} on $\overline{r_c}$ and $\overline{r_p}$ …"

5. p. 13, lines 12-14: The increase of forward scattering with size does not matter much in this case, in which the snow grains are well in the geometric optics regime. Rather, the decrease of albedo with r_{ef} is explained by the fact that the snow extinction (and also scattering) coefficient is inversely proportional to r_{ef}, so that for a given amount of

dust, the single-scattering albedo of the snow-dust mixture is smaller for large snow grains. To put it another way, for larger r_{ef}, solar radiation can penetrate deeper into snow, which increases the chances of absorption by dust.

R: The sentence has been modified as *"the effect of grain radius can be explained by the fact that the snow extinction coefficient is inversely proportional to* r_{ef}*, so that for a given amount of dust, the single-scattering albedo of the snow-dust mixture is smaller for large snow grains."*

6. p. 16, lines 11–25. This discussion on the impact of dust particle effective radius on the ratio of snow broadband albedo for internal vs. external mixing of dust is not valid, for reasons explained in the first major comment. The independence of single scattering properties on dust particle size in the internally mixed cases is an artifact resulting from the use of the Maxwell-Garnett approximation, Eq. (8), which assumes that dust particles are very small compared to the wavelength.

R: We noted that the dust size can influence the albedo of snow-dust mixture for dust-snow external mixing, but not valid for dust-snow internal mixing. Therefore, the uncertainty of broadband snow albedo scaling factor ($E_{\alpha,\ \mathrm{integrated}}$) induced by dust size is entirely caused by the uncertainty of broadband albedo of dust-snow external mixing due to different dust sizes. We have made supplementary explanations for the discussion on the impact of dust particle effective radius on the ratio of snow broadband albedo for dust-snow internal and external mixing, which can be found in p. 17, lines 3-9:

"The results showed that the uncertainty of $E_{\alpha,\ integrated}$ *attributed to dust diameter was ±6.1% for both IDM (uniform) and IDM (central,* $\overline{r_c}$ *<0.75), but it should be emphasized that the uncertainty of* $E_{\alpha,\ integrated}$ *induced by dust sizes comes purely from the uncertainty of broadband albedo of dust–snow external mixing due to different dust sizes. This is because effective complex refractive indices of dust–snow internal mixture are independent of dust particle size (Eq. 8)."*

7. p. 17, lines 25–27. The contrast in the effect of dust on snow albedo between fresh

and aged snow is probably even more pronounced than indicated here, because snow grain effective radius r_{ef} is generally larger for aged snow than fresh snow. In the present calculations, a constant r_{ef} = 200 µm is assumed.

R: We fully agreed with the referee's comments. In order to compare the effects of dust on snow albedo in different snow sampling areas, a constant r_{ef} = 200 µm is assumed in this study, which is comparable to previous observations of fresh snow at mid to high latitudes in winter (Shi et al., 2020; Wang et al., 2017). Therefore, we noted that the effect of dust on snow albedo for aged snow could be underestimated due to the larger snow grain effective radius (r_{ef}) than fresh snow. Details can be found in p. 18, lines 15-18:

"it is worth noting that the effect of dust on snow albedo for aged snow could be underestimated due to the larger snow grain effective radius (r_{ef}) than fresh snow."

Technical and language corrections

1. p. 4, line 11: this should be "Tibetan plateau"?

R: Revised.

2. p. 5, lines 13-14: This is not very clear. Is this what you mean? "...the snow layer can be generally considered semi-infinite in the VIS region if the snow depth is at least 20 cm, and in the near-infrared (NIR) if it is at least 3 cm."

R: This sentence has been rewritten as follows.

"For fairly pure snow, semi-infinite means about 20 cm in the VIS and 3 cm in the near-infrared (NIR) regions of snow depths, respectively."

3. p. 5, line 17: this should be "Kokhanovsky".

R: Revised.

4. p. 10, line 14: "The wavelength of the valley k_{eff}." Do you mean "the wavelength of the minimum of k_{eff}"?

R: Revised to “the wavelength of the minimum of k_{eff}”.

5. p. 11, line 12: “can be regarded as a function of”. Simpler: “depend on”.

R: Revised as suggested.

6. p. 11, lines 16–17: “σ_{abs} increased ... with $\bar{r}_c$ values of 1 to 0.7”. It would be clearer, and probably more correct, to say “...when $\bar{r}_c$ decreased from 1 to 0.7”. There are several other examples like this in the text.

R: We have carefully revised this similar expression throughout the manuscript.

7. p. 13, lines 3-4: “σ_{abs} was decreased by 28%, 32% and 32% ...”. I guess this refers to the difference in σ_{abs} between the uniform case $\bar{r}_c = 1$ and the case $\bar{r}_c < 0{:}75$, but it is not clear from the sentence. Please clarify.

R: This sentence has been rewritten as “σ_{abs} at 500 nm was decreased by 28%, 32%, and 32% from its maximum value (0.08, 0.38, and 3.71 m^{-1}) for dust concentrations of 2, 10, and 100 ppm, respectively, when $\bar{r}_c$ increased from <0.75 to 1.0.”

8. p. 14, line 19: “...such that internal mixing declined more than external mixing”. Presumably this should be “...such that $\alpha_{integrated}$ declined more for internal mixing than external mixing”.

R: Revised as suggested.

9. p. 15, lines 20–24: This sentence can be clarified. “For example, the difference in $E_{\alpha;integrated}$ between dust concentrations of 10 and 20 ppm was 0.011 for IDM (uniform) and 0.015 for IDM (central, $\bar{r}_c < 0{:}75$), while the corresponding differences between dust concentrations of 90 and 100 ppm were only 0.004 and 0.005”.

R: Revised as suggested (See Page 16, Line 4-8).

10. p. 18, line 25–28: This is rather cumbersome. Suggestion: “Therefore, assuming a

completely external mixing of dust and snow grains will underestimate the effects of dust on snow albedo and radiative forcing in numerical models (...). Similarly, assuming completely internal mixing of dust and snow grains will overestimate the effects of dust ...”.

R: Revised as suggested (See Page 20, Line 1-4).

References:

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