Anonymous Referee #1

The authors conducted a theoretical investigation of dust-snow internal mixing and its effects on dust-induced snow albedo reduction. The dust-snow internal mixing has not been quantified by many previous studies, and the present study could potentially increase our understanding of its impact. This manuscript is suitable for this journal and is generally well structured in terms of language. However, I do have one major concern related to the method used in this study. Please see my comments below for details.

R: Thanks for the valuable comments, we have addressed all comments very carefully as detailed below.

Major Comments:

1. My major concern is that the effective medium approximation used by the authors cannot be applied to dust in snow due to the much larger dust size than black carbon (e.g., Bohren, C. 1986: Applicability of effective-medium theories to problems of scattering and absorption by non-homogenous atmospheric particles, J. Atmos. Sci., 43, 468–475). This has also been discussed in detail in Section 2.1 of Flanner et al. (2012, ACP doi:10.5194/acp-12-4699-2012).

R: Although previous literature indicated that the effective medium approximation cannot be applied to dust in snow due to the much larger diameter of dust particles than black carbon (Bohren 1986; Flanner et al., 2012), there are still some studies to solve optically effective complex refractive index of dust composite aerosols based on the effective medium approximation (Cremonesi et al., 2020; Zhang et al., 2020). Besides, we acknowledge that the effective medium approximation used in this study can create some errors for the albedo calculation of dust/snow internal mixing. Therefore, we carefully evaluate the accuracy of our results by comparison to more rigorous calculations found in He et al. (2019), which can be seen in Figure R1 (i.e., Figure 11 in the revised manuscript). More details can be found in Page 19, Lines 1-12 in the revised manuscript as follows.

"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).

Minor Comments:

1. Page 2, Line 10: "absorption coefficient". Did the authors mean the absorption coefficient of snow-dust mixture?

R: To avoid potential ambiguity, the sentence has been revised as "… absorption coefficient and albedo of dust-contaminated snowpack …".

2. Abstract: The authors assumed spherical snow grains and semi-infinite snowpack in their calculations. These need to be mentioned in the abstract to avoid potential confusion.

R: Added as suggested (See Page 2, Line 6 in the abstract).

3. The authors assumed spherical snow grains in this study. But it would be good to discuss the role of snow grain shape in the introduction, because recent studies showed that snow nonsphericity can interact with LAP-snow mixing, which leads to weaker LAP-induced albedo reduction for nonspherical snow shapes than snow spheres (e.g., Dang et al., 2016 JAS: doi:10.1175/JAS-D-15-0276.1; He et al., 2018b JGR, doi:10.1002/2017JD027752). The snow shape can have nontrivial impacts on dust-snow albedo reduction.

R: More descriptions have been added to investigate the role of snow grain shapes in the introduction (See Page 4, Line 2-5).

4. The authors used the effective medium theory to handle dust-snow internal mixing. However, as mentioned in my major comment, this method has its limitation when applying to large particles (e.g., dust) in snow. Some other studies (e.g., Liou et al., 2014, JGR doi:10.1002/2014JD021665; He et al., 2019, JAMES doi:10.1029/2019MS001737) have used another geometric-optics surface-wave approach to deal with dust-snow internal mixing. These two are the pioneering studies

investigating dust-snow internal mixing effects, and need to be discussed in the introduction. Particularly, the authors also need to highlight the difference or novelty of the study compared with these recent studies (e.g., new method, new perspective, etc.?). In the results or discussion section, it is good to also discuss the difference in the results from the present study and the previous study (e.g., He et al., 2019 JAMES) in terms of the impact of dust-snow internal mixing. It would be interesting to see if the results are consistent or not based on two different methods.

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) (see Page 4, Lines 23-28).

"Liou et al. (2014) is the pioneer to investigate the 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."

Besides, we highlighted that the method used in this study greatly simplifies the complexity of snow radiation transfer calculation, and evaluated the effect of non-uniform distribution of dust in snow grains on snowpack absorption coefficient and albedo for the first time. Furthermore, we also carefully access the accuracy of our results by comparison to more rigorous calculations found in He et al. (2019). As shown in Figure R1, 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. More details 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."

5. Page 5, Lines 13-14: Please rephrase this sentence. Did the authors mean 20cm and 3cm of snow depths?

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."

Anonymous Referee #3

The work is well prepared and presented. I recommend accepting it after minor technical corrections.

R: We are very grateful for the referee's positive comments. The followings are our point-by-point responses to the comments. Our responses start with "R:".

1. Figure 1. There are some colour blocks in the figure 1. I don't know the purpose of these blocks. Could the author clarify them?

R: We have added the caption of the color blocks in revised Figure 1.
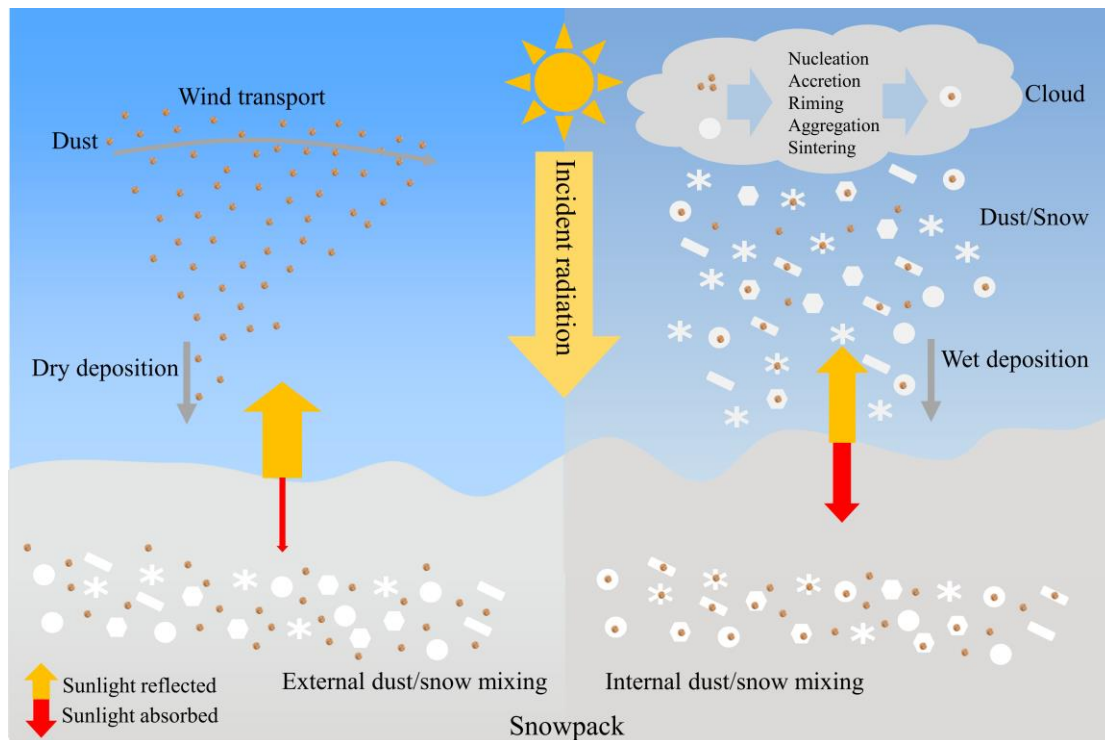
Figure 1. Schematic of dust mixing with snow grains internally. Dust tends to mix externally with snow grains through dry deposition and/or below cloud scavenging, while dust–snow internal mixtures can be produced by nucleation, accretion, riming, aggregation, and sintering during aerosol–cloud–precipitation processes known as wet deposition. Arrows represent how the absorption (red) and reflection (yellow) of incoming sunlight changes with dust–snow mixing state.

2. The Discussion section is too short to be independent. To well balance the structure, I suggest merging this section into conclusion and form a new Discussion and Conclusion section. Otherwise, the authors need to expand it.

R: We fully agreed with the referee's comments. We have reconstructed the discussion section and highlighted the novelty of this study compared with other studies (Liou et al., 2014; He et al., 2019). Meanwhile, we also carefully evaluated the accuracy of our results by comparison to more rigorous calculations found in He et al. (2019). More details can be found in p. 19, lines 1-12.

3. Ming et al. (2016) in figure 9 and 10 were not involving any content in northern

Tibetan Plateau but only Tienshan Mountains. Please check it throughout the manuscript and correct accordingly.

R: We have corrected the inaccurate description and cited another related study by Li et al. (2016).

Anonymous Referee #4

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."

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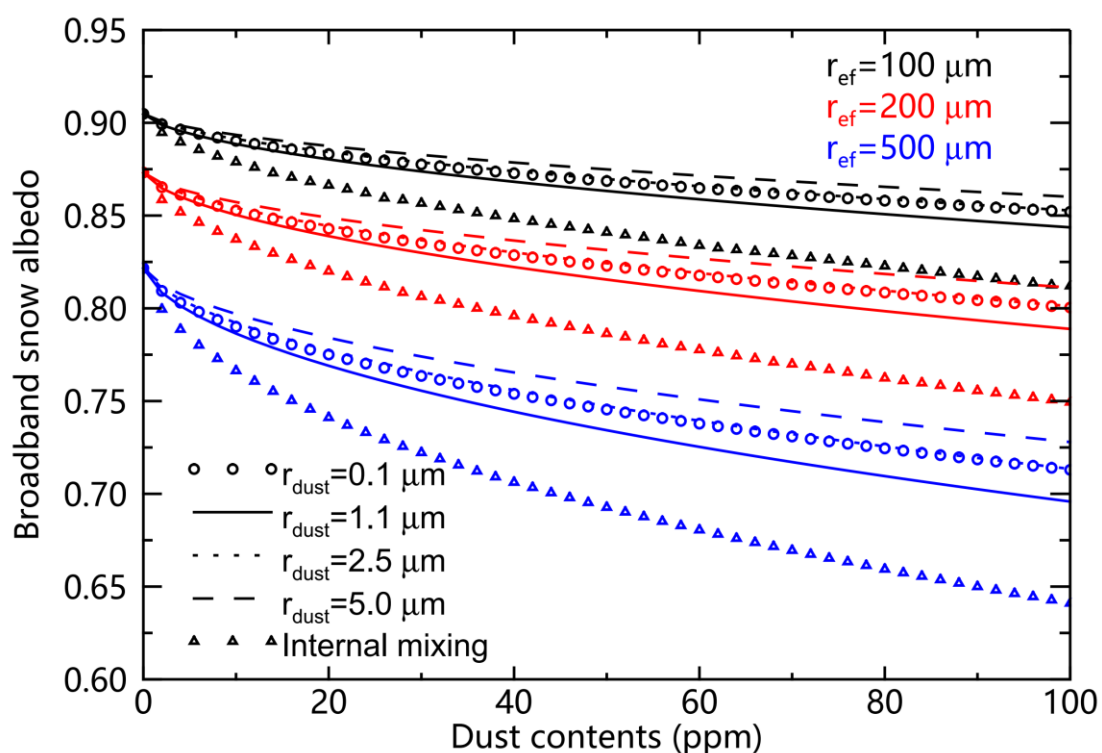
Figure R2. The broadband snow albedo variations with dust contents due to four different dust sizes (0.1, 1.1, 2.5, and 5.0 μm) for dust-snow external mixing, and the results for dust-snow internal mixing were also presented.

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 $\bar{r}_c$ or $\bar{r}_p$?

R: Changed as suggested.

"... monotonic dependence of σ_{abs} on $\bar{r}_c$ and $\bar{r}_p$ ..."

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 ($E_{\alpha,\ integrated}$) 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 $E_{a;integrated}$ 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).

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Enhanced light absorption and reduced snow albedo due to internally mixed mineral dust in grains of snow

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1 **Abstract.** Mineral dust is a major light-absorbing aerosol, which can significantly
2 reduce snow albedo and accelerate snow/glacier melting via wet and dry deposition on
3 snow. In this study, three scenarios of internal mixing of dust in ice grains were
4 analyzed theoretically by combining asymptotic radiative transfer theory and
5 (core/shell) Mie theory to evaluate the effects on absorption coefficient and ~~snow~~
6 albedo of the semi-infinite snowpack consisting of spherical snow grains. In general,
7 snow albedo was substantially reduced at wavelengths of <1.0 μm by internal dust–
8 snow mixing, with stronger reductions at higher dust concentrations and larger snow
9 grain sizes. Moreover, calculations showed that a non-uniform distribution of dust in
10 snow grains can lead to significant differences in the values of the absorption coefficient
11 and ~~snowpack~~ albedo of dust-contaminated snowpack at visible wavelengths relative
12 to a uniform dust distribution in snow grains. Finally, using comprehensive in situ
13 measurements across the Northern Hemisphere, we found that broadband snow albedo
14 was further reduced by 5.2% and 9.1% due to the effects of internal dust–snow mixing
15 on the Tibetan Plateau and North American mountains. This was higher than the
16 reduction in snow albedo caused by black carbon in snow over most North American
17 and Arctic regions. Our results suggest that significant dust–snow internal mixing is
18 important for the melting and retreat of Tibetan glaciers and North American mountain
19 snowpack.

1. Introduction

Snow cover is one of the most reflective surfaces in the Earth system, and plays a crucial role in the atmospheric solar radiation energy budget via snow albedo feedbacks (Di Mauro et al., 2020; Flanner et al., 2011; Jacobson, 2004; Usha et al., 2020; Xie et al., 2018). Previous studies have shown that light-absorbing particles (LAPs) effectively reduce snow albedo and enhance the absorption of solar radiation after deposition, these studies were based on in situ observations and model simulations (Casey et al., 2017; Hadley and Kirchstetter, 2012; Shi et al., 2020; Warren and Wiscombe, 1980; Yasunari et al., 2012; Yasunari et al., 2015). As a result, snow contaminated with LAPs shows significant changes in morphology (Niwano et al., 2012; Rango et al., 1996), chemistry (France et al., 2012; Reay et al., 2012), hydrology (Matt et al., 2018; Qian et al., 2015; Rahimi et al., 2019), snowmelt rate (Kaspari et al., 2015; Warren, 1984;), and radiative properties (Grenfell et al., 2002; Hansen and Nazarenko, 2004; Zhao et al., 2014).

Numerous studies have assessed the potential effects of LAPs, such as black carbon (BC) and mineral dust, on snow albedo by assuming that LAPs mixed outside spherical snow grains (i.e., external mixing) (Flanner et al., 2007; Kokhanovsky, 2013; Libois et al., 2013; Wang et al., 2017; Warren and Wiscombe, 1980). For example, Warren and Wiscombe (1980) calculated snow spectral albedo by solving a radiative transfer equation using Mie theory and δ-Eddington approximations, and found that 10–100 ng g^{-1} of BC and 1–10 µg g^{-1} of dust in old snow decreased albedo by 1%–7% and 2%–10% at 400 nm wavelength, respectively. Flanner et al. (2007) pointed out that the reduction in snow albedo for 1000 ng g^{-1} BC in snow was 0.045 (0.17) with a 50 (1000) µm snow grain radius (R_{ef}) based on the Snow, Ice, and Aerosol Radiation (SNICAR) radiative transfer model. This model utilizes theory from Wiscombe and Warren (1980) and the two-stream radiative transfer solution from Toon et al. (1989). Wang et al. (2017) developed a Spectral Albedo Model for Dirty Snow (SAMDS) based on asymptotic radiative transfer theory, which is a function of the snow grain radius, LAP (e.g., BC, dust) mixing ratios, and the mass absorption coefficients (MACs) of LAPs. Their results

1   revealed that broadband snow albedo decreased 0.03 and 0.003 due to 200 ng g^{-1} of BC

2   and 2 μg g^{-1} of dust in snow, respectively, with a R$_{ef}$ of 200 μm. Additionally, recent

3   studies found that snow nonsphericity can interact with LAP-snow mixing, which leads

4   to weaker LAP-induced albedo reductions for nonspherical snow shapes than snow

5   spheres (e.g., Dang et al., 2016; He et al., 2018, 2019).

6   Recently, direct snowpack observations have shown evidence for the existence of LAP–

7   snow internal mixing (e.g., Horhold et al., 2012; Spaulding et al., 2011). LAPs tend to

8   mix externally with snow grains through dry deposition and/or below cloud scavenging,

9   while internal LAP–snow mixing can be produced by nucleation, accretion, riming,

10   aggregation, and sintering during aerosol–cloud–precipitation processes (i.e., wet

11   deposition; Figure 1) (Flanner et al., 2012). Furthermore, Flanner et al. (2012) found

12   that internal BC/ice mixing (IBM) increased the absorption of snowpack by a factor

13   1.8–2.1 relative to external BC/ice mixing (EBM). He et al. (2018) indicated that IBM

14   enhanced the mean snow albedo reduction over the Tibetan plateau by 30%–60%

15   relative to EBM, based on the updated SNICAR model. Additionally, Dombrovsky and

16   Kokhanovsky (2020) demonstrated that non-uniform BC distribution in ice grains may

17   lead to significantly different absorption coefficients and snowpack albedo in visible

18   (VIS) wavelengths.

19   Numerous studies have addressed the role of IBM in enhancing the absorption of

20   snowpack due to its strong absorption effect relative to other LAPs (Dombrovsky and

21   Kokhanovsky, 2020; Flanner et al., 2012; He et al., 2018; Liou et al., 2011). In contrast,

22   few studies have considered the effects of internal dust/ice mixing (IDM) in snowpack.

23   Liou et al. (2014) is the pioneer to investigate the dust-snow internal mixing effects

24   based on the geometric-optics surface-wave approach. Subsequently, He et al. (2019)

25   used the same method to explicitly quantify the combined effects of dust-snow internal

26   mixing and snow grain nonsphericity on snow optical properties, thereafter, develop a

27   set of new dust-snow parameterizations for land/climate modeling applications for the

28   first time. Actually, dDust particles are generally larger than BC, and act as more

efficient ice nuclei, showing a better ability to influence cloud formation and precipitation (Creamean et al., 2013; Huang et al., 2014). Therefore, they are more likely to mix internally with ice grains. Furthermore, dust can also dominate light absorption and effectively decrease snow albedo because of its relatively high mass abundance (ppm) in snowpack, especially in areas with seasonal and patchy snow cover or mountainous regions (Di Mauro et al., 2015; Gabbi et al., 2015; Painter et al., 2012; Reynolds et al., 2020; Xie et al., 2018). Therefore, it is important to account for IDM when estimating the impact of dust deposition on snow albedo.

In this study, we assess the effects of external/internal mixing of dust with ice grains on the snowpack absorption coefficient and albedo using asymptotic radiative transfer theory and Mie theory. In addition, the uniformity and nonuniformity of dust particle distribution inside ice grains are considered for IDM based on the effective medium approximation, and the combined effects of dust content and snow grain radius on snow albedo are quantified. A schematic of various dust spatial distributions from this study is presented in Figure 2. We further discuss snow albedo sensitivity to complex refractive indices and dust particle size distribution. Based on a comprehensive set of field measurements of dust concentrations, we estimate the reductions in snow albedo by dust external/internal mixing with ice grains across the Northern Hemisphere.

2 Methods

2.1 External mixing model

For fairly pure snow, semi-infinite means semi-infinite is generally defined as absorptions of about 20 cm in the VIS and 3 cm in the near-infrared (NIR) regions of snow depths, respectively (Zhou et al., 2003). For a semi-infinite snow layer under diffuse illumination conditions, albedo can be calculated using an asymptotic analysis of radiative transfer theory, which is valid in small absorptions (Kokhanovsky and Zege, 2004; Zege et al., 1991):

$$\alpha_\lambda = exp\left(-4S_\lambda\right) \tag{1}$$

1   where α_λ is the spectral snow albedo, λ is the wavelength, S_λ is the similarity

2   parameter, and

3    $$S_\lambda = \sqrt{\frac{\sigma_{abs}}{3\sigma_{ext}(1-g)}} \qquad (2)$$

4   In Eq. (2), σ_{abs} and σ_{ext} are the absorption and extinction coefficients, respectively,

5   and g is the asymmetry parameter (the average cosine of the phase function of the

6   medium).

7   According to Eq. (18) and (25) in Kokhanovsky and Zege (2004), the extinction

8   coefficients of particles can be expressed as:

9    $$\sigma_{ext} = \frac{\cancel{l_{tr}}1}{l_{tr}(1-g)\cancel{1-g}} = \frac{3C_v}{2r_{ef}} \qquad (3)$$

10   where l_{tr} is the photon transport path length, $C_v = \rho_{snow}/\rho_{ice}$ is the volumetric

11   snow particle concentration, and the values $\rho_{ice} = 916.7$ kg m^{-3} and $\rho_{snow} = 300$ kg

12   m^{-3} are used in subsequent calculations. r_{ef} is the effective snow grain radius, which

13   is equal to the radius of the volume-to-surface equivalent sphere ($r_{ef} = \frac{3\overline{V}}{4\overline{A}}$) where $\overline{V}$

14   and $\overline{A}$ are the average volume and cross-sectional (geometric shadow) area of snow

15   grains, respectively.

16   For external dust/ice mixing (EDM) in a dust-contaminated snowpack, the total

17   absorption coefficient (σ_{abs}) can be derived from the absorptions by snow (σ_{abs}^{snow}) and

18   dust (σ_{abs}^{dust}):

19   $$\sigma_{abs} = \sigma_{abs}^{snow} + \sigma_{abs}^{dust} \qquad (4)$$

20   For example, consider a hypothetical case of snow composed of monodispersed,

21   spherical grains of ice. Although non-spherical snow grains lead to a slight increase in

22   snow albedo, Dang et al. (2016) found that the albedo of a snowpack consisting of non-

23   spherical snow grains can be mimicked by using smaller, spherical grains; thus, we do

24   not consider the effect of non-spherical snow grains in this study. Therefore, we used

25   the following equation for the absorption coefficient of snow (Dombrovsky and Baillis,

26   2010):

27   $$\sigma_{abs}^{snow} = \frac{0.75 C_v \cdot Q_{abs}^{ice}}{r_{ef}} \qquad (5)$$

1   where Q_{abs}^{ice} is the efficiency factor of absorption for a single ice grain, and the value

2   of Q_{abs}^{ice} can be calculated for homogeneous spherical ice grains considered in classical

3   Mie theory.

4   The absorption coefficient of dust (Aoki et al., 2000; Marley et al., 2001; Warren et al.,

5   2006) is expressed as:

6    $$\sigma_{abs}^{dust} = \frac{Q_{abs}^{dust} \cdot \pi \cdot \left(r_{ef}^{dust}\right)^2}{\frac{4}{3}\pi \cdot \left(r_{ef}^{dust}\right)^3} \cdot C_{dust} = \frac{3Q_{abs}^{dust}}{4r_{ef}^{dust}} \cdot C_{dust} = MAC_{abs}^{dust} \cdot \rho_{dust} \cdot C_{dust} \qquad (6)$$

7   where Q_{abs}^{dust} and MAC_{abs}^{dust} is the absorption efficiency and MAC of dust,

8   respectively, that can be obtained via Mie theory, and ρ_{dust} and r_{ef}^{dust} represent the

9   density and effective dust radius, respectively. In this study, ρ_{dust} was assumed to be

10   2500 kg m^{-3} (Zender et al., 2003). We also assumed a log-normal dust size distribution

11   with a geometric mean diameter of 0.65 μm and standard deviation of 2.0 (equivalent

12   to an effective radius of 1.1 μm), which represents dust from large-scale transport

13   (Formenti et al., 2011; Maring et al., 2003) that is likely smaller in size than from local

14   soil (Kok, 2011). The effects of dust size on snow optical properties and albedo were

15   further quantified through sensitivity simulations (see section 3.4). Dust volumetric

16   concentrations (C_{dust}) are expressed as:

17    $$C_{dust} = \frac{\rho_{snow} \cdot C_{dust}^*}{\rho_{dust}} \qquad (7)$$

18   where C_{dust}^* is the mass concentration of dust in snow (kg kg^{-1}). Thus, the spectral

19   albedo of dust-contaminated snow for EDM can be easily calculated with Eq. (1) to (7).

20   **2.2 Internal mixing model**

21   For the IDM (Figure 2), we first determined the effective optical constants of ice

22   containing small dust particles via an effective medium approximation (Maxwell-

23   Garnett and Larmor, 1904). According to this approach, the complex permittivity of a

24   composite medium in ice grains can be calculated in terms of particle polarizability by

25   applying the Lorentz-Lorenz formula (Koledintseva et al., 2009; Markel, 2016). We

26   used the following relationships to calculate effective complex refractive indices (RIs),

1      $m_{ef} = n_{ef} - ik_{ef}$ at known values of $m_{ice} = n_{ice} - ik_{ice}$ for pure ice and $m_{dust} = n_{dust} - ik_{dust}$

2   for dust:

3      $$m_{ef}^2 = m_{ice}^2 \frac{2\delta_{dust}(m_{dust}^2 - m_{ice}^2) + m_{dust}^2 + 2m_{ice}^2}{2m_{ice}^2 + m_{dust}^2 - \delta_{dust}(m_{dust}^2 - m_{ice}^2)} \tag{8}$$

4   where δ_{dust} is the local dust fraction volume in an ice grain. We obtained the spectral

5   complex RIs of ice from Warren and Brandt (2008) and the spectral complex RIs of

6   dust from Dang et al. (2015). The imaginary part of the complex RIs of ice (k_{ice}) and

7   dust (k_{dust}) associated with absorption is shown in Figure 3. We also evaluated the effect

8   of dust on the imaginary part of the effective complex RIs (k_{ef}) assuming dust mass

9   concentrations (C_{dust}^*) of 1–100 ppm (or μg g^{-1}) in snow.

10   In all variations of the spatial distribution of dust particles in snow, the dust mass

11   concentration was assumed to be constant, which means that the local dust fraction

12   volume may differ. In an example of spherical ice grains with uniformly distributed

13   dust, the dust fraction volume in an ice grain was determined as:

14      $$\delta_{dust}^0 = \frac{C_{dust}^*}{C_v} \frac{\rho_{ice}}{\rho_{dust}} \tag{9}$$

15   where the ratio of C_{dust}^*/C_v is the mass fraction of dust in the ice grain.

16   We considered two cases of non-uniform dust distributed in a spherical ice grain with

17   radius r_{ef}: (1) We assumed that the same mass of dust is uniformly distributed in the

18   central part of the ice grain ($r_{ef}^{dust} < r_c \leq r_{ef}$). (2) We assumed all of the dust was in

19   the surface layer of the ice grain ($r_{ef}^{dust} < r_p \leq r_{ef}$) (Figure 2). In both cases the local

20   value of δ_{dust} increases as:

21      $$\delta_{dust} = \delta_{dust}^0 \Big/ \psi \qquad \psi = \begin{cases} \bar{r}_c^3 & \text{central pollution} \\ 1 - (1 - \bar{r}_p)^3 & \text{peripheral pollution} \end{cases} \tag{10}$$

22   where $\bar{r}_c = r_c/r_{ef}$ and $\bar{r}_p = r_p/r_{ef}$. $\bar{r}_c = 1$ and $\bar{r}_p = 1$ correspond to uniformly

23   distributed dust when $\psi = 1$ and $\delta_{dust} = \delta_{dust}^0$, and $\psi < 1$ and $\delta_{dust} > \delta_{dust}^0$ in

24   other cases. Obviously, dust particles increase the imaginary part of the effective

25   complex RIs in polluted ice grains (Figure 3).

26   In summary, the m_{ef} of a spherical ice grain with uniformly and non-uniformly

distributed ~~dust can be calculated,~~ according to Eq. (8) to (10), then their corresponding absorption efficiencies can be obtained using classical Mie theory and core/shell Mie theory, respectively. The spectral snow albedo for IDM can be easily calculated using Eq. (1) to (5).

2.3 Broadband snow albedo calculations

The spectral albedo (α_λ) is integrated over the solar spectrum ($\lambda = 300$–2500 nm) and weighted by incoming solar irradiance (E_λ) to calculate broadband snow albedo (Hadley and Kirchstetter, 2012):

$$\alpha_{integrated} = \frac{\int \alpha_\lambda E_\lambda d\lambda}{\int E_\lambda d\lambda} \tag{11}$$

Following the study of Dang et al. (2017), the incoming solar irradiance we used 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 (Pu et al., 2019). The SBDART model is a widely used atmospheric radiation transfer model based on a collection of highly developed physical models, including the Discrete Ordinate Radiative Transfer module (Stamnes et al., 1988), low-resolution atmospheric transmission models, and Mie theory. The SBDART model can be used to compute radiative transfer at different heights and directions under both clear and cloudy sky conditions. Details on the SBDART model can be found in Ricchiazzi et al. (1998).

2.4 Dust concentration measurements

To estimate the effect of dust on snow albedo in real snowpack, we collected a comprehensive set of in situ dust concentration measurements during field campaigns to Inner Mongolia, China (IMC) (Wang et al., 2013), the Tibetan Plateau (TP) (~~Ming et al., 2016;~~ Li et al., 2017, 2018; Li et al., 2016, 2019; Niu et al., 2017; Qu et al., 2014; Zhang et al., 2017, 2018), Sapporo, Japan (SJ) (Kuchiki et al., 2015), the European Alps (EP) (Di Mauro et al., 2015; Lim et al., 2014), and North American mountains (NAM) (Painter et al., 2012; Reynolds et al., 2020). The field campaigns conducted in the TP

1 can be further grouped into three regions, as in previous $\delta^{18}O$ precipitation studies (Yao

2 et al., 2013). These three distinct domains were associated with the Indian monsoon

3 (Southern TP), westerlies (Northern TP), and transition (Central TP) (Yao et al., 2013).

4 It is worth noting that we only considered regions with higher dust concentrations (>1

5 ppm), such that polar regions are not included in this study. Measurements of dust in

6 the snow samples were generally obtained by weighing the filter before and after

7 filtration using a microbalance.

8 **3. Results**

9 **3.1 Impact on the imaginary part of the effective complex RIs**

10 We evaluated the effect of dust on k_{ef}, including k_{ice} and k_{dust} associated with absorption

11 (Figure 3). The k_{dust} was in a narrow range (~0.001–0.01) and gradually decreased with

12 increasing wavelength in the ultraviolet (UV) and VIS regions, then remained stable in

13 the NIR band. The k_{ice} varied by eight orders of magnitude from the UV (~10^{-11}) to NIR

14 (~10^{-3}) bands, and increased with increasing wavelength, except at 1.03 μm where k_{ice}

15 decreased slightly as a result of the presence of ice absorption features (Warren, 2019).

16 Figure 3 also shows the k_{ef} with dust mass concentrations ranging from 1 to 100 ppm

17 and wavelengths of 300–1500 nm. The k_{ef} clearly varied depending on the wavelength

18 and increased with dust mass concentrations. For a given dust mass concentration, k_{ef}

19 decreased with wavelengths from UV to VIS, then increased from VIS to NIR. For

20 example, the value of k_{ef} decreased from 4.26×10^{-8} at 300 nm to 1.36×10^{-8} at 500

21 nm, then rose to 1.73×10^{-6} at 1000 nm at a dust concentration of 10 ppm. Moreover,

22 the wavelength of ~~the minimum of~~the valley k_{ef} ~~value~~ varied from ~500 nm to ~650 nm

23 depending on the dust mass concentrations (1 to 100 ppm). Additionally, it is worth

24 noting that k_{ef} was not sensitive to dust mass concentrations in wavelengths >1000 nm,

25 which was generally consistent with k_{ice}, because the difference between k_{dust} and k_{ice} is

26 more than compensated by the much larger difference in ice and dust concentration ~~the~~

27 ~~order of magnitude of k_{ice} was comparable to k_{dust}~~ at those wavelengths. Conversely, k_{ef}

1   showed significant differences relative to k_{ice} in the UV and VIS regions, with higher

2   dust mass concentrations demonstrating larger differences. For example, k_{ef} was

3   enhanced by 3, 21, and 205 times at 500 nm relative to k_{ice} for dust mass concentrations

4   of 1, 10, and 100 ppm, respectively.

5   **3.2 Impact on spectral snow absorption coefficient and albedo**

6   Dust in snow effectively enhances the snow absorption coefficient, but its effect on the

7   snow asymmetry factor and extinction efficiency is negligible (He et al., 2019).

8   Therefore, we mainly focused on the effects of EDM and three cases of IDM (uniform,

9   central, and peripheral) on the snow absorption coefficient (σ_{abs}). Figure 4a displays the

10   σ_{abs} for EDM and IDM (uniform) as a function of wavelength at different dust

11   concentrations. We used a snow grain radius of 200 μm (Figure 4), which is comparable

12   to previous observations of seasonal snow at mid to high latitudes in winter (Shi et al.,

13   2020; Wang et al., 2017). The results showed that EDM and IDM have distinct impacts

14   on σ_{abs} in UV and VIS, but small effects at wavelengths >1000 nm, which is due to

15   the optical properties of snow being affected by LAPs in UV and VIS and primarily

16   affected by snow itself at wavelengths >1000 nm. Additionally, σ_{abs} increased with

17   increased dust mass concentrations. For instance, σ_{abs} increased from 0.007 m^{-1} (pure

18   snow) to 0.03, 0.14, and 1.37 m^{-1} at 500 nm with 2, 10, and 100 ppm of dust with EDM,

19   respectively. For IDM (uniform), σ_{abs} increased to 0.06, 0.28, and 2.80 m^{-1} at 500 nm

20   with 2, 10, and 100 ppm of dust, respectively, with corresponding enhancement factors

21   of σ_{abs} ($E_{\sigma_{abs}}$, defined as the absorption coefficient of IDM divided by EDM) were

22   1.84, 2.00, and 2.05. Furthermore, the σ_{abs} for two cases of non-uniform dust

23   distribution in a spherical ice grain (r_{ef} = 200 μm) ~~can be regarded as a function~~

24   ~~of~~depends on the wavelength, dust mass concentrations, $\bar{r_c}$, and $\bar{r_p}$ (Figure 4b and 4c).

25   We note that $\bar{r_c}$ and $\bar{r_p}$ values of 1 correspond to uniformly distributed dust, and the

26   σ_{abs} increases and decreases (with the decrease of $\bar{r_c}$ and $\bar{r_p}$) for IDM (central) and

27   IDM (peripheral), respectively. For example, σ_{abs} increased by 29%, 32%, and 33%

28   (500 nm) at dust mass concentrations of 2, 10, and 100 ppm, respectively, when $\bar{r_c}$

1 decreased from 1 to 0.7~~with $\bar{r}_c$ values of 1 to 0.7~~ for IDM (central). However, σ_{abs}

2 decreased by 41%, 44%, and 44% (500 nm) at dust mass concentrations of 2, 10, and

3 100 ppm, respectively, when $\bar{r}_p$ decreased from 1 to~~with $\bar{r}_p$ of 1 to~~ 0.1 for IDM

4 (peripheral). This indicates that the IDM (central) further enhanced snowpack light

5 absorption compared with the IDM (uniform), while the IDM (peripheral) reduced

6 snowpack light absorption with a corresponding σ_{abs} between the value of σ_{abs} for

7 IDM (uniform) and EDM. Figure 4d–f quantitatively shows the spectral snow

8 absorption coefficient enhancement for IDM ($E_{\sigma_{abs}}$). The enhancement decreased

9 sharply with increasing wavelengths, then reduced to 1.0 (i.e., no enhancement) at

10 wavelengths longer than ~1.0 μm because of strong dust absorption and weak snow

11 absorption at shorter wavelengths. Obviously, $E_{\sigma_{abs}}$ was affected by dust mass

12 concentration, $\bar{r}_c$, and $\bar{r}_p$, but $E_{\sigma_{abs}}$ was insensitive to dust mass concentration at

13 wavelengths <450 nm.

14 Figure 5a–c shows the spectral snow albedo (α_λ) for EDM and IDM; α_λ~~IDM~~ was

15 consistent with σ_{abs}, with the effects mainly present at wavelengths <1000 nm.

16 Generally, α_λ decreased with increased dust mass concentrations in UV and VIS, and

17 IDM was shown to further trigger the reduction of α_λ. For example, for EDM α_λ was

18 ~0.97, 0.95, 0.85 (at 500 nm) for dust concentrations of 2, 10, 100 ppm, respectively,

19 which was higher than the values for IDM (uniform) (~0.96, 0.93, 0.79, respectively).

20 Compared with IDM (uniform), the α_λ for IDM (central) decreased by 0.5%, 1.1%,

21 and 3.5% (at 500 nm) for dust concentrations of 2, 10, and 100 ppm, respectively, when

22 $\bar{r}_c$ decreased from 1 to 0.7~~with $\bar{r}_c$ values of 1 to 0.7~~. α_λ for IDM (peripheral)

23 increased by 0.8%, 1.9%, and 6.2% (at 500 nm) for the same dust concentrations, when

24 $\bar{r}_p$ decreased from ~~with $\bar{r}_p$ from~~ 1 to 0.1. Moreover, the wavelength of the maximum

25 α_λ value varied from ~500 nm to ~650 nm depending on the dust mass concentrations,

26 which is consistent with changes of k_{ef}. Figure 5d–f shows the ratio (E_{α_λ}) of snow

27 spectral albedo for IDM to EDM where we observed that the E_{α_λ} increased with

28 increasing wavelengths and dust concentrations, and then became stable at 1.0. This is

1 because IDM can enhance the light absorption of snowpack more effectively at shorter

2 wavelengths and higher dust concentrations (Figure 4). Additionally, the values of $\bar{r}_c$

3 and $\bar{r}_p$ also have non-negligible effects on E_{α_λ}, which can be decreased and increased

4 with decreasing $\bar{r}_c$ and $\bar{r}_p$, respectively.

5 We found that the optical properties of an ice grain containing uniformly distributed

6 dust in its center, or concentric surface layer, can be affected by $\bar{r}_c$ or $\bar{r}_p$. To better

7 understand this effect, Figure 6a–b displays the σ_{abs} at 500 nm as a function of $\bar{r}_c$ and

8 $\bar{r}_p$ with different dust concentrations and r_{ef}. This demonstrates r_{ef} has negligible

9 effects on σ_{abs} due to the geometric optical limits at $r_{ef} \approx 50$ μm, which shows the

10 universal (independent of r_{ef}) monotonic dependence of σ_{abs} on $\bar{r}_c$ and $\bar{r}_p$ for ice

11 grains with $r_{ef} \geq 50$ μm (Velesco et al., 1997). As a result, the spectral absorption

12 coefficient of snow containing polydispersed ice grains can be obtained using our

13 results for a monodispersed model. Interestingly, σ_{abs} did not depend on $\bar{r}_c$ when $\bar{r}_c$

14 <0.75 and decreased almost linearly at higher $\bar{r}_c$ values (Figure 6a); this phenomenon

15 can be explained by geometric optical effects (Mackowski et al., 1990). However, σ_{abs}

16 was significantly affected by the dust mass concentration; for example, σ_{abs} at 500 nm

17 was decreased by 28%, 32%, and 32% from its maximum value (0.08, 0.38, and 3.71

18 m^{-1}) for dust concentrations of 2, 10, and 100 ppm, respectively, ~~with~~ when $\bar{r}_c$

19 increased from <0.75 to 1.0. The monotonic increase in σ_{abs} with the relative thickness

20 of the polluted ice grain surface layer (i.e., $\bar{r}_p$) was also noteworthy (Figure 6b). The

21 core/shell Mie theory calculations for ice grains with a thin surface layer ($\bar{r}_p = 0.01$)

22 gave almost the same σ_{abs} as that obtained for the EDM. As a result, the σ_{abs}

23 increased rapidly with $\bar{r}_p$ <0.4 and then increased more slowly until $\bar{r}_p = 1$, which

24 corresponds to IDM (uniform).

25 The α_λ at 500 nm as a function of $\bar{r}_c$ and $\bar{r}_p$ with different dust concentrations and

26 r_{ef}, is illustrated in Figure 6c and 6d. In general, α_λ at 500 nm decreased with

27 increasing dust mass concentration and r_{ef}; the effect of grain radius can be explained

28 by the fact that the snow extinction coefficient is inversely proportional to r_{ef}, so that

1 for a given amount of dust, the single-scattering albedo of the snow-dust mixture is

2 smaller for large snow grains~~increasing the forward scattering with grain size~~ (Gardner

3 and Sharp, 2010). For a given dust mass concentration and r_{ef}, the α_λ at 500 nm

4 increased from its minimum value with $\bar{r}_c$ <0.75 to the maximum value with $\bar{r}_c$ = 1,

5 corresponding to the findings of IDM (uniform). For example, at dust concentrations

6 of 2, 10, and 100 ppm, and a fixed r_{ef} of 100 μm, the α_λ at 500 nm increased by 0.2%,

7 0.7%, and 0.8%, respectively, when $\bar{r}_c$ increased ~~with $\bar{r}_c$~~ from <0.75 to 1. When the

8 r_{ef} was fixed at 500 μm, the α_λ at 500 nm increased by 1.9%, 2.5%, and 6.2% at dust

9 concentrations of 2, 10, and 100 ppm, respectively. Conversely, the α_λ at 500 nm

10 decreased from its maximum value when $\bar{r}_p$ = 0.01 (similar to EDM) to the minimum

11 value with $\bar{r}_p$ = 1, corresponding to the case of IDM (uniform). For example, the α_λ

12 at 500 nm decreased by 0.6%, 1.4%, and 1.4% when~~with~~ $\bar{r}_p$ increased from 0.01 to 1

13 for dust concentrations of 2, 10, and 100 ppm, respectively, and a fixed r_{ef} of 100 μm,

14 whereas for a r_{ef} of 500 μm, α_λ decreased by 3.3% (2 ppm dust), 4.7% (10 ppm), and

15 10.1% (100 ppm). These results indicate that dust mass concentrations and r_{ef} can

16 amplify the influence of $\bar{r}_c$ or $\bar{r}_p$ on snow albedo. Moreover, the effect of dust mass

17 concentration on snow albedo is similar to r_{ef}. For example, dust mass concentrations

18 of 10 and 100 ppm and r_{ef} of 100 and 50 μm gave similar α_λ at 500 nm to dust mass

19 concentrations of 2 and 10 ppm and r_{ef} of 500 μm. According to this result, spectral

20 albedo measurements at a single wavelength are insufficient to obtain the mass fraction

21 of dust in snow cover because the same effect can also be explained by a combination

22 of different ice grain sizes and a non-uniform distribution of dust inside the grains. It

23 means that additional information is needed to determine accurate dust mass

24 concentrations. This may be a set of measurements at various wavelengths in the VIS

25 and NIR spectral ranges.

26 **3.3 Effects on broadband snow albedo**

27 Compared with the spectral optical properties, broadband results can provide more

28 general knowledge for the relevant research community. Figure 7 shows the spectrally

weighted α_λ ($\alpha_{integrated}$) over 300–2500 nm of a typical surface solar spectrum at mid

to high latitudes, which is comparable with previous studies (Dang et al., 2017; Wang

et al., 2017). Because the results of IDM (peripheral) effects on snow albedo fell

between results from EDM and IDM (central), we do not consider the case of IDM

(peripheral) in the following discussion. Instead, we focus on the effects of dust mass

concentration and r_{ef} on broadband snow albedo for EDM and IDM (uniform, central).

Similar to α_λ, $\alpha_{integrated}$ generally decreased with increasing dust mass

concentrations and r_{ef} such that $\alpha_{integrated}$ declined more for internal mixing ~~declined~~

~~more~~ than external mixing. $\alpha_{integrated}$ showed ranges of 0.60–0.92, 0.54–0.92, and

0.51–0.92 for EDM, IDM (uniform), and IDM (central, $\bar{r}_c$ <0.75), respectively, with

dust mass concentrations of 0–100 ppm and r_{ef} of 50–1000 μm. For a given dust mass

concentration and r_{ef}, $\alpha_{integrated}$ for IDM (uniform) was smaller than EDM, which is

due to higher light absorption in the UV and VIS bands for IDM (uniform) relative to

EDM (Figure 4a). While $\alpha_{integrated}$ for IDM (uniform) was larger compared with

IDM (central, $\bar{r}_c$ <0.75), this can be attributed to the fact that radiation is focused near

the center of an ice grain with IDM (central) (Ackerman and Toon, 1981; Bohren, 1986),

enabling further absorptions from inclusions near the center of a grain due to the lensing

effect (Mackowski et al., 1990). For example, $\alpha_{integrated}$ (dust concentration of 20

ppm, r_{ef} of 500 μm) was 0.73 for IDM (uniform), less than EDM of 0.76, but higher

than IDM (central, $\bar{r}_c$ <0.75) of 0.72.

To quantify the effects of IDM on broadband snow albedo relative to EDM, we defined

a broadband snow albedo scaling factor ($E_{\alpha,\ integrated}$), which refers to the ratio of

$\alpha_{integrated}$ of IDM to EDM. Generally, for dust mass concentrations from 0 to 100 ppm

and r_{ef} of 50–1000 μm, $E_{\alpha,\ integrated}$ varied from 0.89 to ~1.00 for IDM (uniform) (Figure

8a) and from 0.85 to ~1.00 for IDM (central, $\bar{r}_c$ <0.75) (Figure 8b). $E_{\alpha,\ integrated}$

decreased significantly with increasing dust mass concentration and r_{ef}. In addition, $E_{\alpha,}$

$_{integrated}$ for IDM (central, $\bar{r}_c$ <0.75) was smaller than IDM (uniform). These results

have implications for the effects of IDM in real environments. For example, IMC has

1   typical dust concentrations of ~10 ppm (Wang et al., 2013), so $E_{\alpha,\text{ integrated}}$ (r_{ef} of 50–

2   1000 µm) was 0.96–0.99 and 0.95–0.99 for IDM (uniform) and IDM (central, $\overline{r}_c$ <

3   0.75), respectively. In contrast, dust concentrations are typically ~100 ppm in the TP

4   (Ming et al., 2016; Li et al., 2017, 2018), so $E_{\alpha,\text{ integrated}}$ ranged from 0.89 to 0.98 and

5   0.85 to 0.96 for IDM (uniform) and IDM (central, $\overline{r}_c$ <0.75), respectively. The results

6   show that IDM (uniform) and IDM (central, $\overline{r}_c$ <0.75) reduced broadband snow

7   albedo by ~2.5% and ~3.0%, respectively, in clean snow and ~6.5% and ~9.5%,

8   respectively, in polluted snow relative to EDM. Moreover, the sensitivity of $E_{\alpha,\text{ integrated}}$

9   to mineral dust decreased with increasing dust concentrations. For example, the

10   difference in $E_{\alpha,\text{ integrated}}$ (r_{ef} of 500 µm) between dust concentrations of 10 and 20 ppm

11   was 0.011 and 0.015 for IDM (uniform) and IDM (central, $\overline{r}_c$ <0.75), respectively,

12   while the corresponding differences between dust concentrations of 90 and 100 ppm

13   were only 0.004 and 0.005. ~~when dust concentrations were 10 20 ppm, but only 0.004~~

14   ~~and 0.005 for IDM (uniform) and IDM (central, $\overline{r}_c$ <0.75), respectively, when dust~~

15   ~~concentrations were 90 100 ppm.~~ These results provide a convenient method to

16   calculate the albedo of IDM when the albedo of EDM has been obtained for a given

17   dust mass concentration and r_{ef}.

18   **3.4 Uncertainties**

19   Although we calculated the imaginary RI values of dust using previous studies (section

20   2.2), there are still large variations which strongly depend on dust composition (e.g.,

21   hematite/iron content) (Balkanski et al., 2007; Wagner et al., 2012). To roughly account

22   for this, we estimated the influence of chosen imaginary RI values on spectrally

23   weighted snow albedo ($E_{\alpha,\text{ integrated}}$) by increasing and decreasing the calculated

24   imaginary RI values by 50%. These changes in imaginary RIs are plausible because

25   they are consistent with other studies (McConnell et al., 2010; Wagner et al., 2012).

26   The results showed that $E_{\alpha,\text{ integrated}}$ uncertainties attributed to the imaginary RIs of dust

27   were ±3.9% and ±5.2% for IDM (uniform) and IDM (central, $\overline{r}_c$ <0.75), respectively.

28   In contrast, observations have displayed large variations in the size distribution of dust

1 in the atmosphere and snow, and this variation is strongly affected by the dust source

2 and transport (Mahowald et al., 2014; Shao et al., 2011). In our standard simulation, we

3 assumed a log-normal dust size distribution with a geometric mean diameter of 0.65

4 μm and a standard deviation of 2.0 (equivalent to an effective radius of 1.1 μm), which

5 is typical for dust transported long-range (Formenti et al., 2011; Maring et al., 2003);

6 nearer sources of dust tend to be larger (Kok, 2011). Therefore, we investigated the

7 effects of dust particle size on our results by assuming another two log-normal size

8 distributions with effective radii of 2.5 μm and 5.0 μm, which were within the observed

9 size ranges in the atmosphere and snow and comparable with previously analyzed dust

10 particle sizes (Maring et al., 2003; Shao and Mao, 2016; Zhang et al., 2003). The results

11 showed that the uncertainty of $E_{\alpha,\,integrated}$ attributed to dust diameter was ±6.1% for both

12 IDM (uniform) and IDM (central, $\overline{r_c}$ <0.75), but it should be emphasized that the

13 uncertainty of $E_{\alpha,\,integrated}$– induced by dust sizes comes purely from the uncertainty of

14 broadband albedo of dust–snow external mixing due to different dust sizes. This is

15 because effective complex refractive indices of dust–snow internal mixture effective

16 optical constants of ice containing small dust particles (i.e., internal mixing) are

17 independent of dust particle size (Eq. 8). Overall, the total uncertainty of $E_{\alpha,\,integrated}$

18 from variations of imaginary RIs and dust diameter was ±11.0% and ±11.2% for IDM

19 (uniform) and IDM (central, $\overline{r_c}$ <0.75), respectively.

20 **3.5 Measurement-based estimate of the effects of dust on snow albedo**

21 Finally, widespread dust concentrations in snow across the Northern Hemisphere were

22 obtained to assess the effects of dust on snow albedo in real snowpack. Figure 9 shows

23 measured dust concentrations in snow in different regions; dust concentrations spanned

24 a broad range of values because of spatial and temporal variations in emissions,

25 transportation, and deposition among the different regions. Dust concentrations widely

26 varied from ~3 ppm to ~600 ppm, with the highest concentration in NAM and lowest

27 in the EP (Di Mauro et al., 2015; Lim et al., 2014; Painter et al., 2012; Reynolds et al.,

28 2020). However, snow samples collected in the days after a significant dust transport

1   event showed that dust concentrations in snow can be up to ~70 ppm in the EP (Di

2   Mauro et al., 2015). Additionally, the average dust concentrations in fresh snow were

3   18, 6, and 28~~17~~ ppm in the southern TP, central TP, and northern TP, respectively,

4   similar to the IMC (12 ppm) (Wang et al., 2013). However, dust concentrations in aged

5   snow (120, 300, and 140 ppm) were one to two orders of magnitude higher than in fresh

6   snow, indicating the important correlation between snow type and dust concentration

7   (Zhang et al., 2017, 2018).

8   We calculated the broadband snow albedo for EDM, IDM (uniform), and IDM (central,

9    $\bar{r}_c$ <0.75) based on the measured dust concentrations (Figure 10). The results showed

10   that broadband snow albedo decreased by 0.8%, 1.4%, and 1.6% in the EP for EDM,

11   IDM (uniform), and IDM (central, $\bar{r}_c$ <0.75), respectively, which was similar to SJ.

12   However, the broadband snow albedo decreased by up to 5.6%, 8.1%, and 9.4% in the

13   EP after a significant dust transport event, indicating strong snow albedo reduction

14   during these events. In addition, broadband snow albedo was reduced by 2.0%, 3.1%,

15   and 3.6% in IMC for EDM, IDM (uniform), and IDM (central, $\bar{r}_c$ <0.75), respectively.

16   Similar results were also found for the southern TP, central TP, and northern TP where

17   the broadband snow albedo for fresh snow was reduced by 2.5%, 1.4%, and ~~2.5~~3.3%,

18   respectively, for EDM, 3.9%, 2.1%, and ~~3.8~~5.1% for IDM (uniform), and 4.5%, 2.4%,

19   and ~~4.3~~5.7% for IDM (central, $\bar{r}_c$ <0.75). However, the broadband snow albedo was

20   more significantly reduced for aged snow: up to 6.0%, 8.1%, and 7.5% for EDM, 9.5%,

21   11.6%, and 10.5% for IDM (uniform), and 10.9%, 13.2%, and 12.3% for IDM (central,

22    $\bar{r}_c$ <0.75) in the southern, central, and northern TP, respectively. This indicates that the

23   effects of dust on snow albedo showed stronger reductions during snowmelt periods, it

24   is worth noting that the effect of dust on snow albedo for aged snow could be

25   underestimated due to the larger snow grain effective radius (r_{ef}) than fresh snow.

26   Moreover, the largest broadband snow albedo reductions were found in NAM with

27   ranges of 9.8%–17.6%, 13.9%–24.1%, and 15.9%–27.0% for EDM, IDM (uniform),

28   and IDM (central, $\bar{r}_c$ <0.75), respectively. These results suggest that the effects of

external or internal dust–snow mixing on snow albedo are particularly significant for the TP and NAM regions, with stronger reductions in albedo. Therefore, these results can have significant impacts on both local hydrological cycles and regional climate change (Oaida et al., 2015; Xie et al., 2018).

4. Discussion

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.

Over the past few decades, the effects of dust in snow on reductions in albedo ~~has~~ have been widely demonstrated (Skiles et al., 2018; Zhang et al., 2018). However, the magnitude of these effects has only been studied in a few regions, and uncertainties still remain. Our study indicates that the albedo of dust-contaminated snowpack can be affected by the dust–ice mixing state. In particular, IDM enhanced light absorption and reduced snow albedo more significantly compared with EDM. For example, in IMC and the TP, IDM reduced snow albedo by ~5% relative to EDM at a typical dust mass concentration of 20 ppm and a snow grain radius of 500 μm. This exceeds the

contribution of BC to snow light absorption over most areas of North America and the Arctic (Dang et al., 2017). In addition, the effects of IDM on snow albedo were amplified by higher dust mass concentrations and larger snow grain sizes. We therefore strongly suggest that IDM must be considered in future climate models, particularly to more accurately evaluate the climate in areas where snowpack is heavily contaminated with dust and is experiencing melting.

The mixed state between dust and snow gradually progresses from partial external mixing to wholly internally mixed. 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 (e.g., Dang et al., 2015; Nagorski et al., 2019). Similarly, assuming completely internal mixing of dust and snow grains will overestimate the effects of dust (e.g., He et al., 2019; Liou et al., 2014). Therefore, information gained solely from the external mixing of dust and snow grains will underestimate the effects of dust on snow albedo and radiative forcing in numerical models (e.g., Dang et al., 2015; Nagorski et al., 2019). Similarly, only using information from internal mixing of dust and snow grains will overestimate the effects of dust on snow albedo and radiative forcing (e.g., He et al., 2019; Liou et al., 2014). Zhao et al. (2014) underestimated the effects of dust by treating wet-deposited dust as externally mixed with snow grains. In future studies, we recommend the actual ratio between external and internal mixing for dust in snow be examined with an environmental scanning electron microscope equipped with a cold stage.

5. Conclusions

In this study, the effects of dust particles on absorption coefficients and snow albedo were theoretically analyzed by combining asymptotic radiative transfer theory and (core/shell) Mie theory. We initially considered external mixing – when dust is present between ice grains – and variations of internal mixing of dust within ice grains. We found that snow spectral absorption coefficients of IDM were larger than EDM across UV to NIR wavelengths, but were negligible at wavelengths >1000 nm. The absorption

enhancement (relative to EDM) was wavelength-dependent and increased with increased dust concentrations.

Compared with a uniform distribution of dust particles in ice grains, our calculations showed that non-uniformly distributed dust particles may lead to significantly different snow spectral absorption coefficients in the VIS band. Snow spectral absorption coefficients were further increased when all of the dust was positioned in the central part of ice grains, while the maximum absorption coefficient was found when the radius of a dust-polluted core was <75% of the ice grain radius. In contrast, snow spectral absorption coefficients decreased when all of the dust was positioned in the surface layer of ice grains, and the minimum absorption coefficient was observed in the thin surface layer of dust-polluted ice grains, which was similar for EDM. As a result, broadband snow albedo decreased by up to 21%, 30%, and 33% for EDM, IDM (uniform), and IDM (central, $\bar{r}_c < 0.75$), respectively, at dust concentrations of 100 ppm and r_{ef} of 1000 μm.

Based on comprehensive field measurements across the Northern Hemisphere, the effect of dust on snow albedo in real snowpack was evaluated by assuming external and internal dust–snow mixing. The largest reductions in broadband snow albedo were in NAM because that region had the highest average dust concentrations; IDM (uniform) and IDM (central, $\bar{r}_c < 0.75$) further decreased snow albedo by 4.6%–7.8% and 6.8%–11.4%, respectively, compared with EDM. This implies an important influence of internal dust–snow mixing in NAM.

Data availability

The code of (core/shell) Mie theory used in this study can be found at http://gwest.gats-inc.com/software/software_page.html.

Author contributions

WX designed the study and evolved the overarching research goals and aims. STL wrote the first draft with contributions from all co-authors. STL and CJC applied formal

techniques such as statistical, mathematical, and computational to analyze study data. CY and XXY collected the dust measurements across the Northern Hemisphere. ZY and PW provided the majority of the methodology and software. CQL and ZXL provided technical guidance. All authors contributed to the improvement of results and revised the final paper.

Competing interests

The authors declare that they have no conflict of interest.

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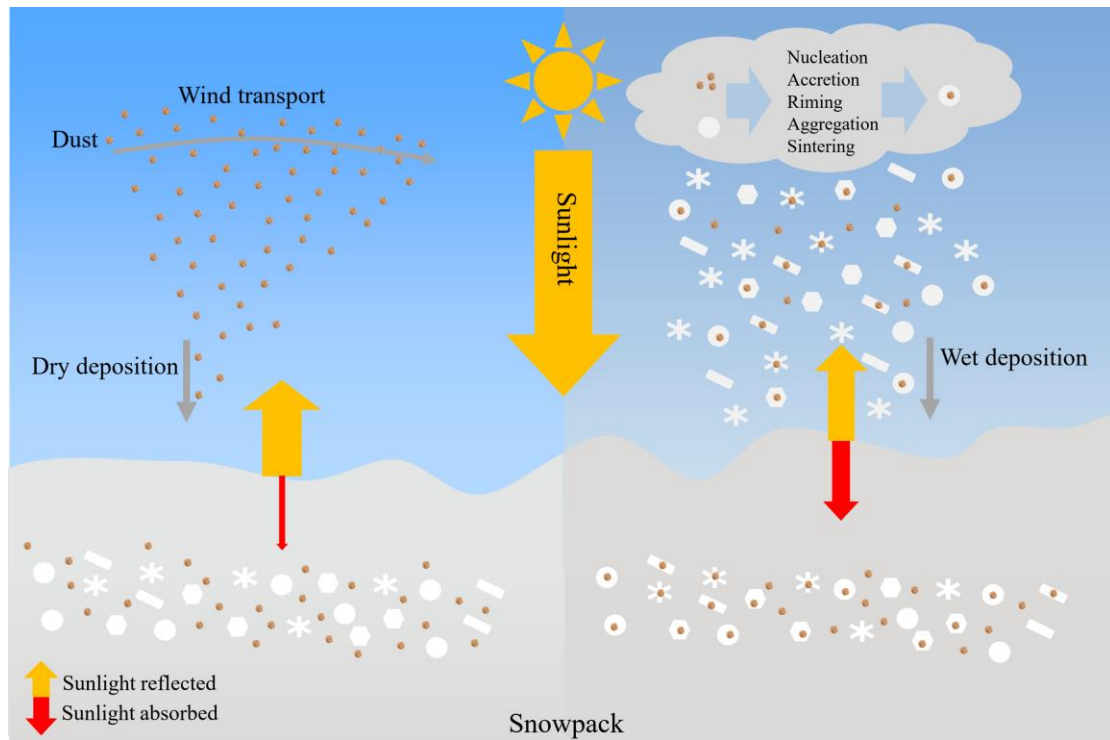
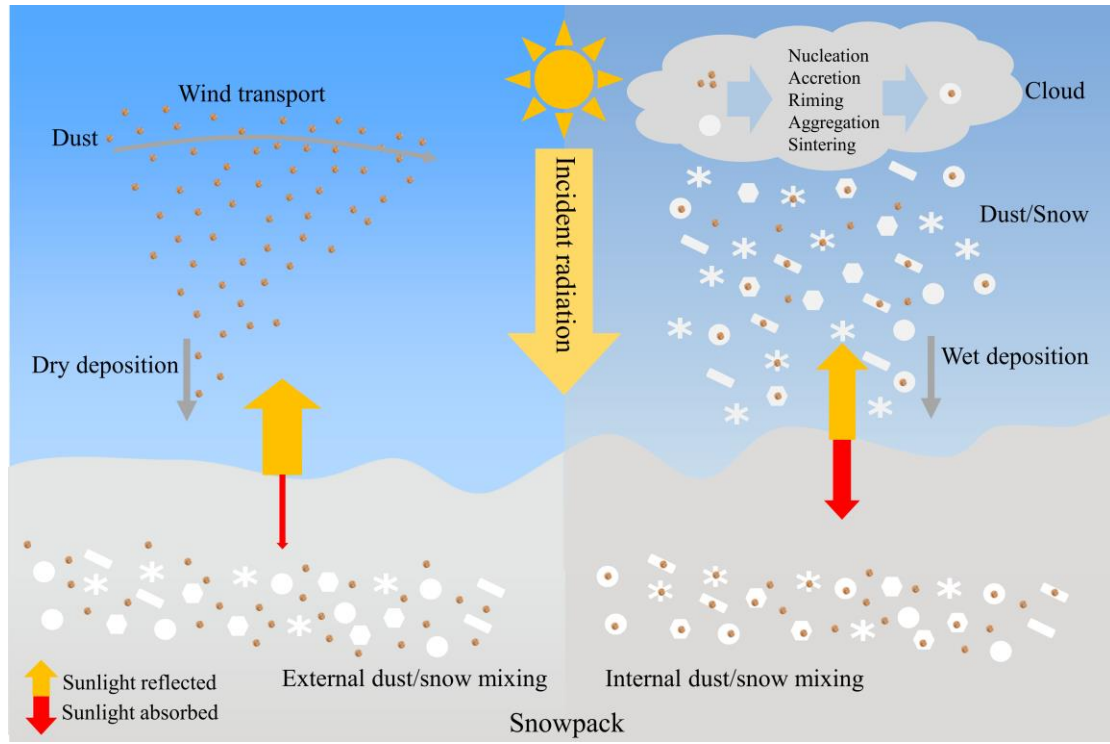
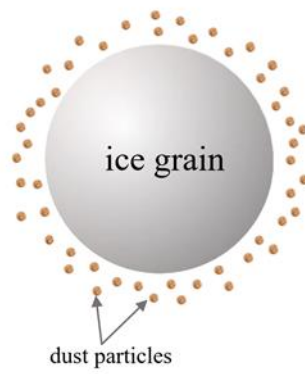
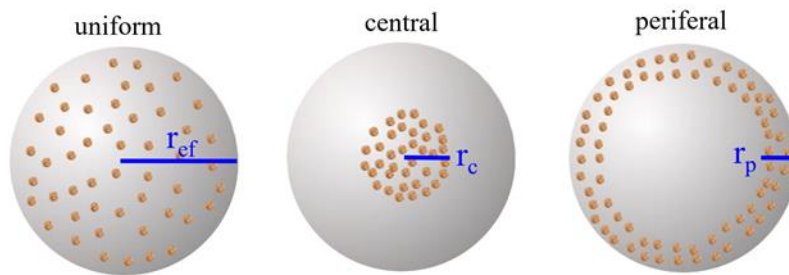
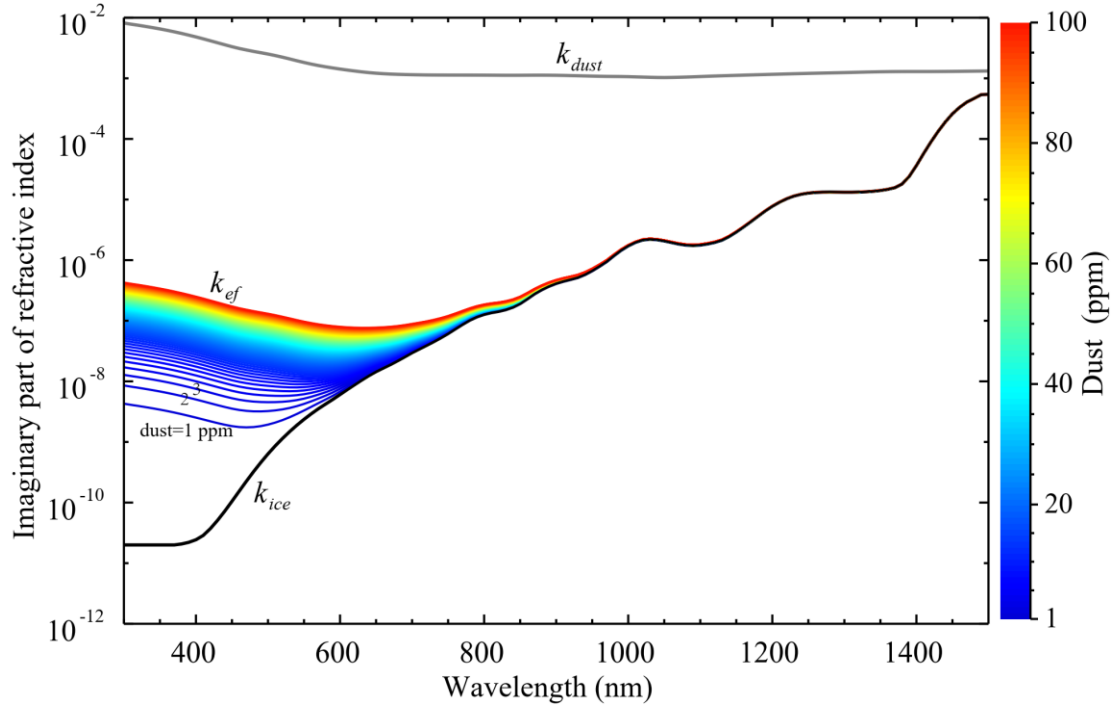


Figure 1. Schematic of dust mixing with snow grains internally. Dust tends to mix externally with snow grains through dry deposition and/or below cloud scavenging, while dust–snow internal mixtures can be produced by nucleation, accretion, riming, aggregation, and sintering during aerosol–cloud–precipitation processes known as wet deposition. Arrows represent how the absorption (red) and reflection (yellow) of incoming sunlight changes with dust–snow mixing state.

External dust/ice mixing (EDM)

Internal dust/ice mixing (IDM)

2   **Figure 2.** Schematic depicting various mixing scenarios of snow grains and dust
3   particles.

2 **Figure 3.** Imaginary part of the spectral complex refractive indices of ice (k_{ice}) and dust
3 (k_{dust}) (Warren and Brandt, 2008; Dang et al., 2015), with the imaginary part of the
4 effective complex refractive indices (k_{ef}) as a function of wavelength, at dust mass
5 concentrations (C_{dust}^{*}) of 1–100 ppm (or µg g^{-1}) in snow.

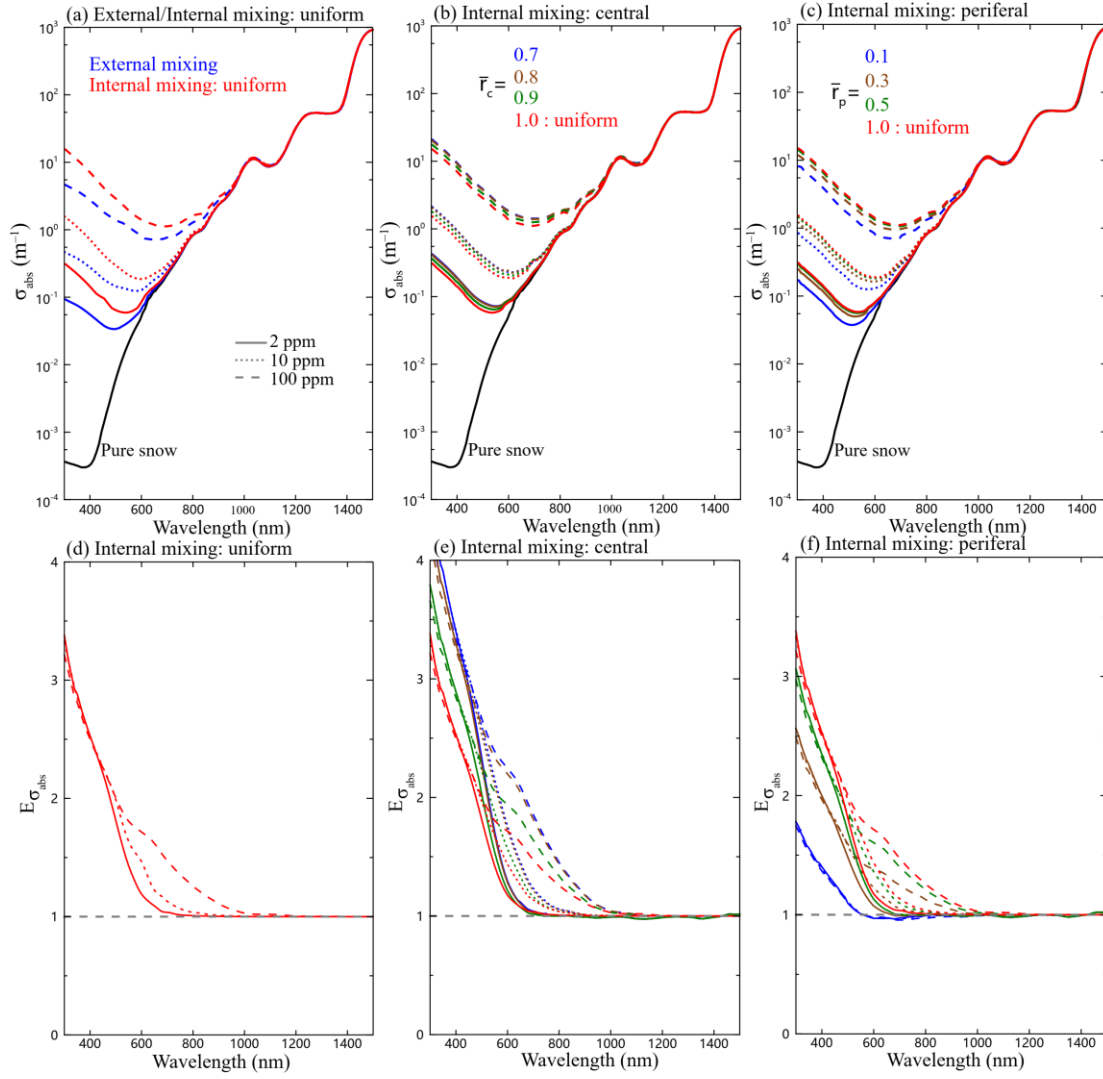



Figure 4. Snow absorption coefficients (σ_{abs}) for dust–snow (a) external and internal mixing (uniform), (b) internal mixing (central), and (c) internal mixing (peripheral), as a function of wavelength with different dust concentrations and $\overline{r_c}$ and $\overline{r_p}$. The corresponding enhancement ($E_{\sigma_{abs}}$) caused by (d) internal mixing (uniform), (e) internal mixing (central), and (f) internal mixing (peripheral) relative to external mixing, is shown as a function of wavelength. The snow grain radius was assumed to be 200 µm.

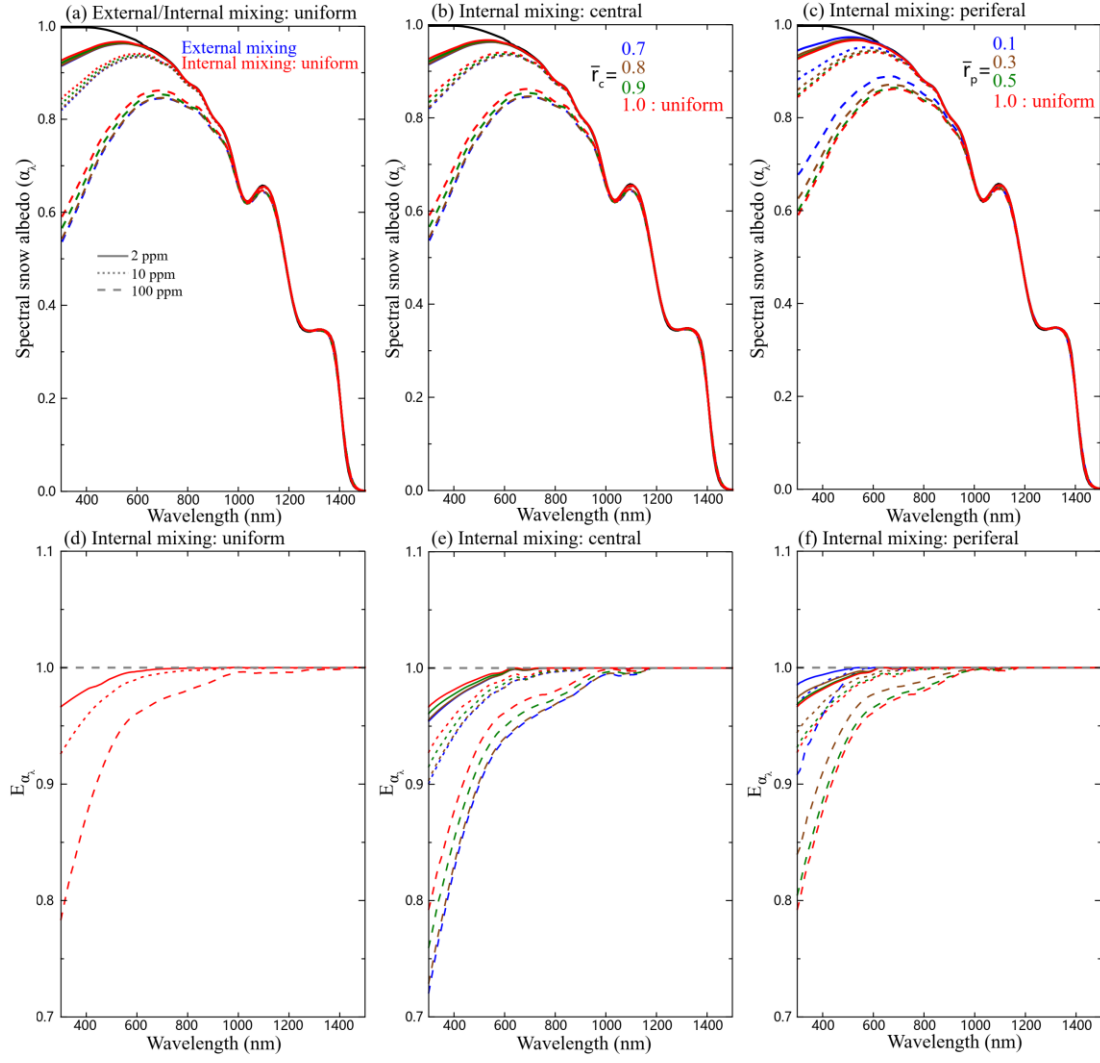
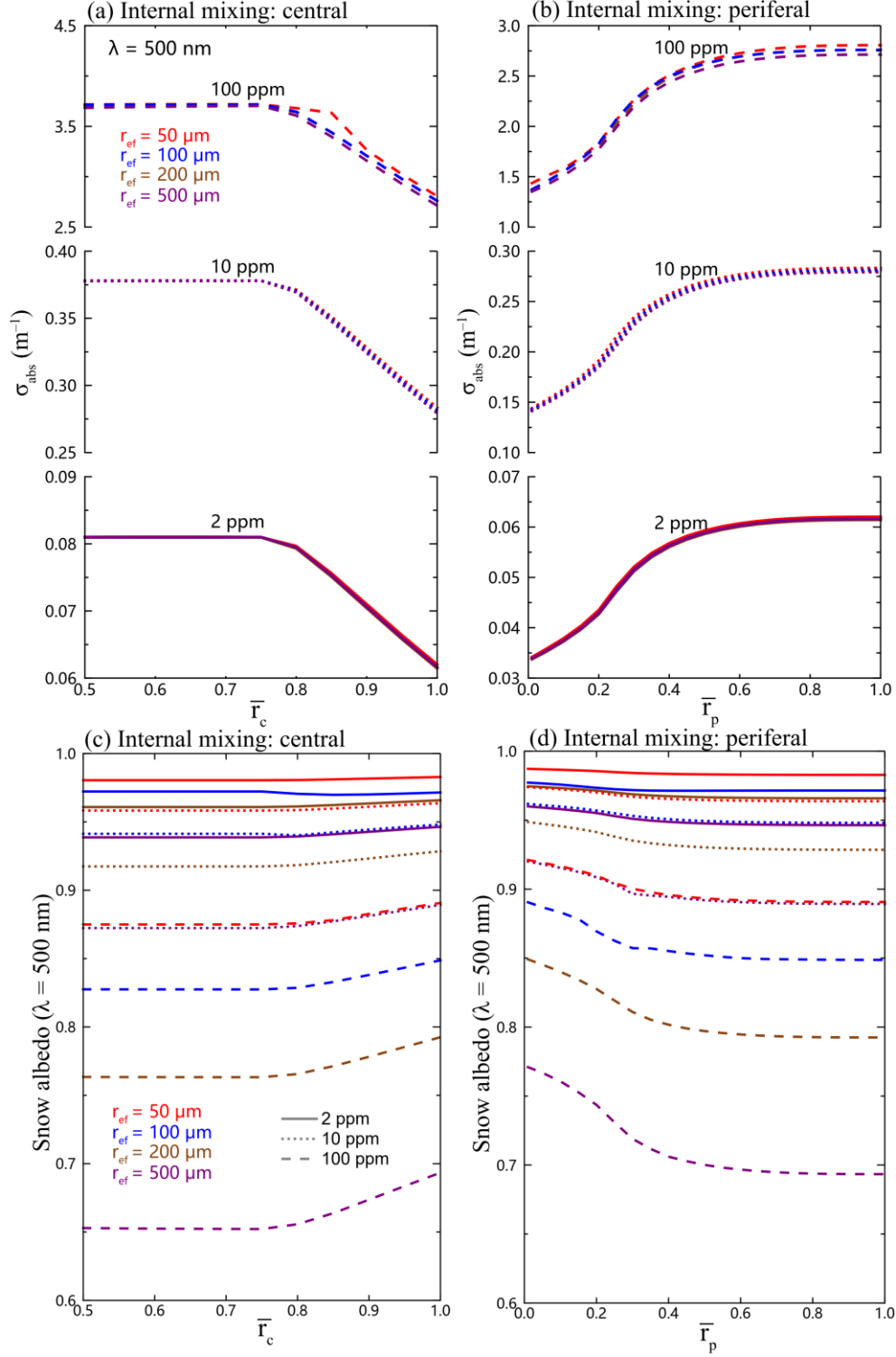
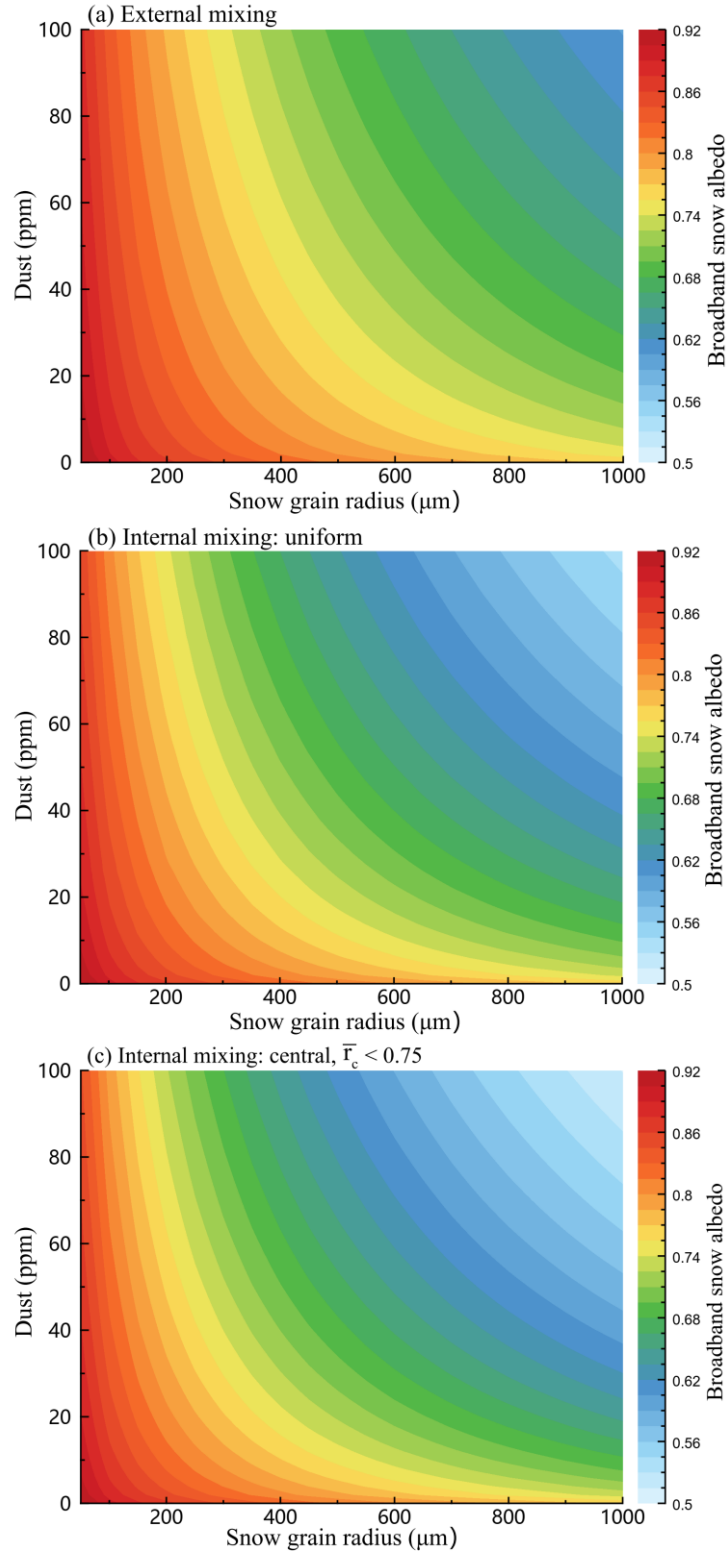



Figure 5. Same as Figure 4, but for spectral snow albedo (α_λ).

2 **Figure 6.** The snow absorption coefficient (σ_{abs}) at 500 nm wavelength as a function of

3 $\overline{r_c}$ and $\overline{r_p}$ for (a) internal mixing (central) and (b) internal mixing (peripheral) with

4 different snow grain radii and dust mass concentrations. (d) and (e) are the same as (a)

5 and (b), but for snow albedo at 500 nm wavelength.

2 **Figure 7.** Broadband snow albedo ($\alpha_{\text{integrated}}$) variations affected by different dust
3 mass concentrations and snow grain radii for (a) external mixing, (b) internal mixing
4 (uniform), and (c) internal mixing (central, $\overline{r}_c$ <0.75).

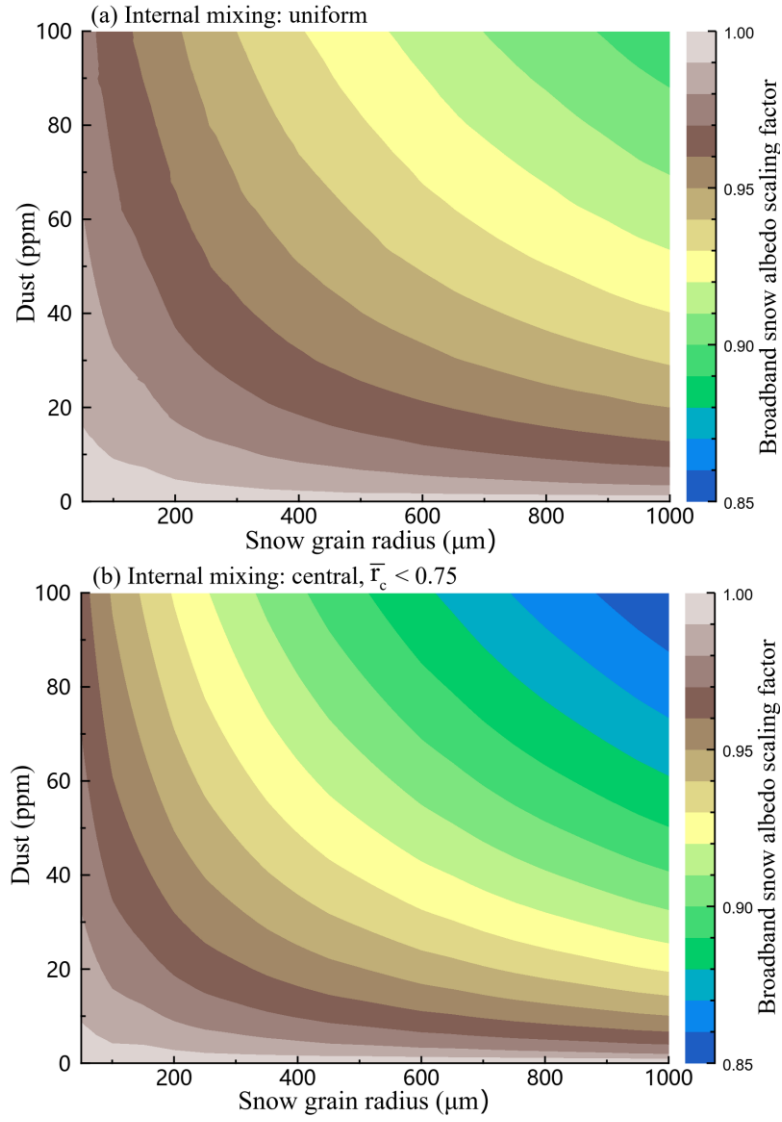



Figure 8. Variations in the broadband snow albedo scaling factor ($E_{\alpha,\text{integrated}}$, ratio of $\alpha_{\text{integrated}}$ for IDM to EDM) due to different dust mass concentrations and snow grain radii for (a) internal mixing (uniform) and (b) internal mixing (central, $\overline{r}_c < 0.75$).

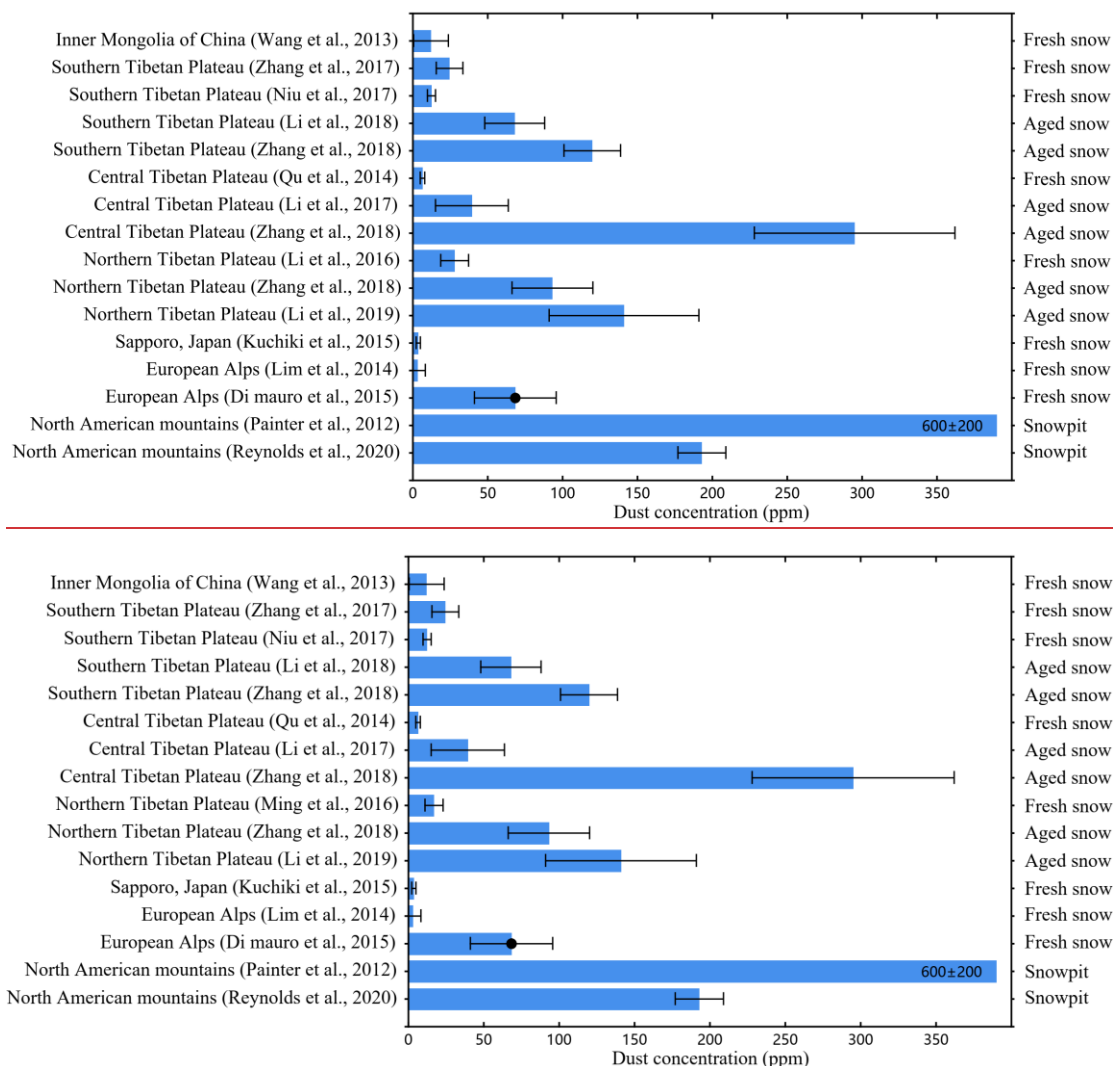



Figure 9. In situ measurements of dust concentrations in snow (fresh snow, aged snow, and snowpit from field sampling in different regions of the Northern Hemisphere. The solid black circle represents snow samples that were collected days after a significant dust transport event.

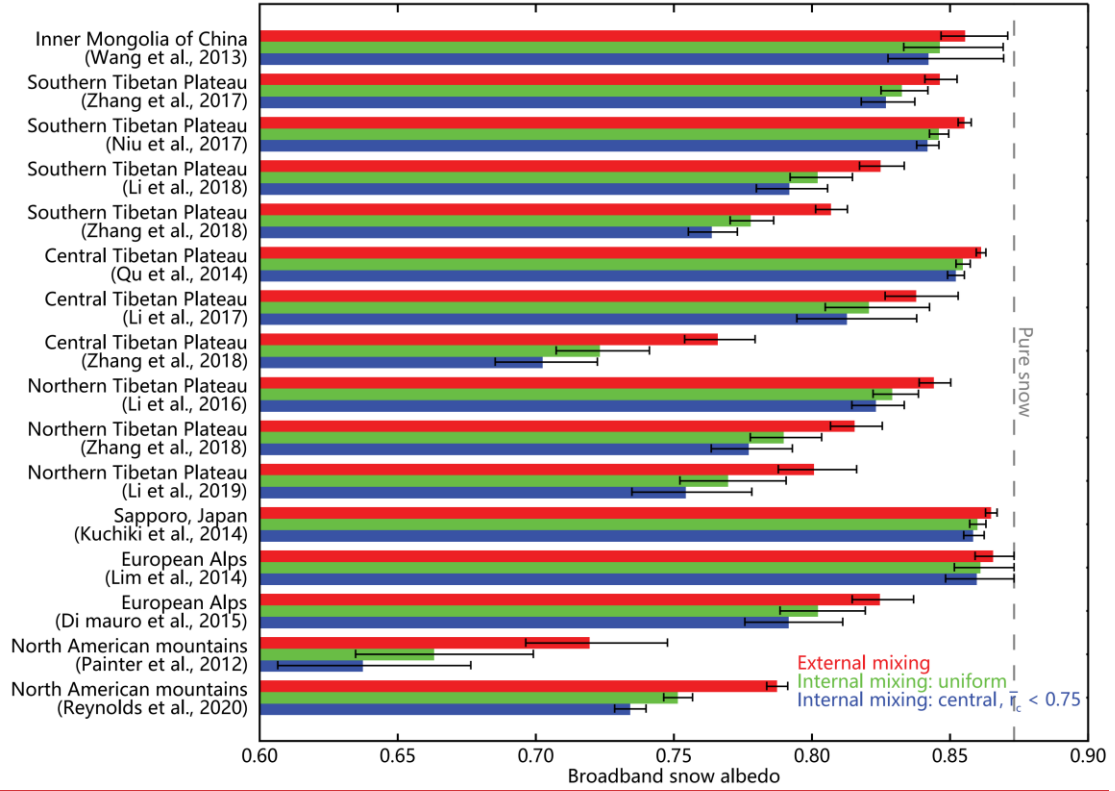
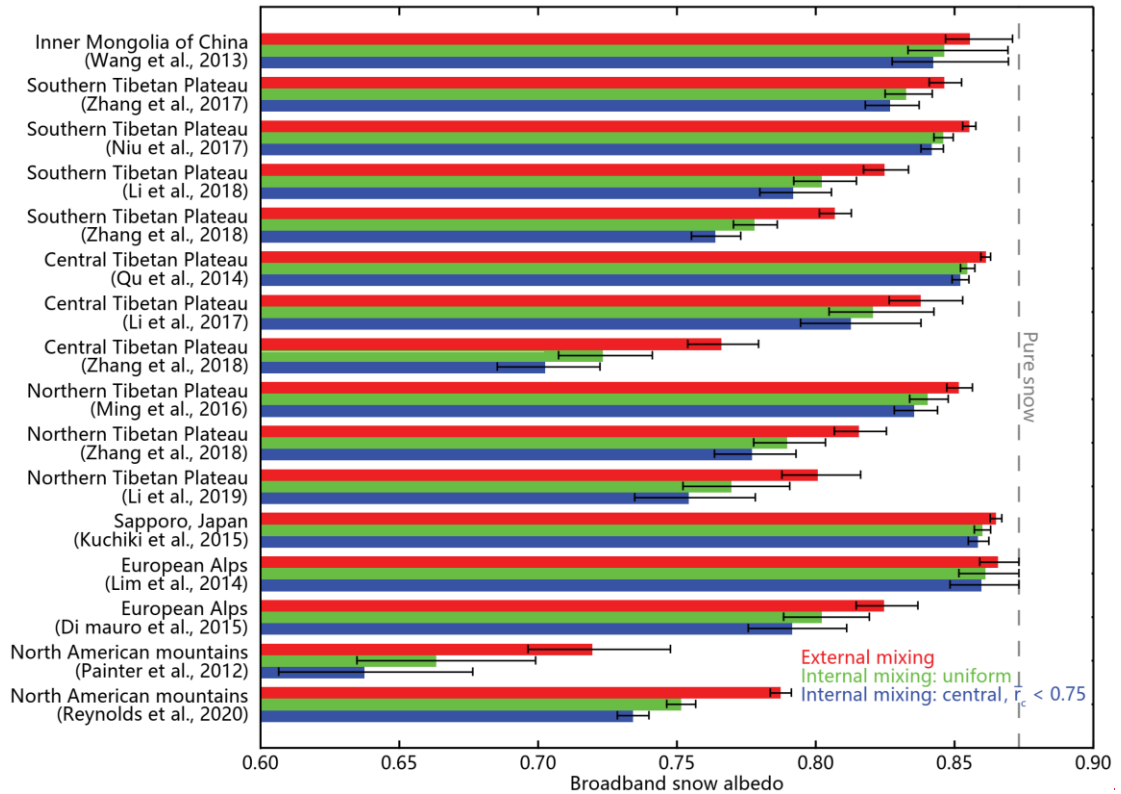



Figure 10. Calculated broadband snow albedo based on dust concentration measurements in different areas for dust–snow external mixing, internal mixing (uniform), and internal mixing (central, $\bar{r}_c$ <0.75). The dashed line represents broadband albedo of pure snow, and the snow grain radius was assumed to be 200 μm.

2 **Figure 11.** Comparisons of snow albedo reduction (Δα) under the cloudy sky caused

3 by dust-snow uniform internal mixing (y-axis) and external mixing (x-axis) for this

4 study (blue) and He et al. (2019) (red).