

Editor: Nikos Hatzianastassiou

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Thank the Editor very much for handling the manuscript. We take into account all the comments from referees and make revisions. Please check the response to the referees and the revised manuscript.

Anonymous Referee #1

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

General comments

I thank the authors for their substantial efforts in revising the manuscript. Most of my original comments have been addressed satisfactorily. However, a few minor points should still be addressed, along with some technical/language corrections.

R: Thank you very much for the positive comments, which will encourage us to do more in-depth research in the future. Moreover, the referee's comments and suggestions are quite significant that can help us to improve the paper quality substantially. We have addressed all of the comments carefully according to the suggestions. All of the detailed responses can be seen as follow.

Specific comments

1. p. 2, line 13: I think it should be clarified what the stated value (2.9 W m^{-2}) actually represents. E.g., "This value represents land areas with complete or near-complete snow cover, with little or no vegetation above the snow."

R: We have added the detailed description about what " 2.9 W m^{-2} " actually represents as "...with daily radiative forcing ($RF_{MODIS,daily}^{LAPs}$) values of $\sim 2.9 \text{ W m}^{-2}$, over land areas with complete or near-complete snow cover, with little or no vegetation above the snow in Northern Hemisphere." in p. 2, line 13-14 as suggestion.

2. p. 13, line 16: For Eq. (2) to be correct, E_{dif}^{clear} should be the diffuse spectral irradiance on a *horizontal surface* and E_{dir}^{clear} the direct spectral irradiance on a *surface perpendicular to the sun*. Please state this in the text, and importantly, check that this is indeed what SBDART provides.

R: We have added the statement about E_{dif}^{clear} and E_{dir}^{clear} as “... $E_{dif}^{clear}(\lambda; \varphi)$ denote the diffuse spectral irradiance on a horizontal surface and $E_{dir}^{clear}(\lambda; \varphi)$ denote the direct spectral irradiance on a surface perpendicular to the sun...” in p. 13, line 17-19. And we determine that the direct and diffuse spectral irradiance are indeed provided by SBDART just like Painter et al. (2012).

3. p. 25, line 16: “significant altitude-dependent” trend? This requires a bit more explanation, e.g. are the values increasing or decreasing with altitude in the Russian Arctic?

R: Revised as suggestion in p. 25, line 16-17: “In Russian Arctic, $\Delta\alpha_{MODIS,corrected}^{LAPs}$ and $RF_{MODIS,daily}^{LAPs}$ values increase with altitude of $\sim 0.012-0.048$ and $\sim 1.0-7.3 \text{ W m}^{-2}$.”

4. p. 28, lines 4–18: It seems to me that the first factor listed here might be the best candidate for explaining why the ratio $\Delta\alpha_{MODIS,daily}^{LAPs}/\Delta\alpha_{in-situ,daily}^{LAPs}$ to be larger than 1, especially for relatively clean snow (the last three factors also cause errors, but it is not obvious whether they usually give rise to an overestimate or underestimate). Any vegetation in the MODIS scene likely reduces the derived albedo, and this probably also applies to the effect of snow surface roughness (Manninen et al.: Effect of small-scale snow surface roughness on snow albedo and reflectance, The Cryosphere Discussions, <https://doi.org/10.5194/tc-2020-154>, in review, 2020.). To my understanding this cannot be accounted for in the pure snow albedo calculation with SNICAR, which might give rise to a positive albedo bias compared to that derived from MODIS — yielding therefore an overestimate of the albedo reduction attributed to LAPs?

R: As the referee said, the effect of snow surface roughness and vegetation, which were without regarding in SNICAR, probably reduce the derived albedo from MODIS and therefore result in overestimate of the albedo reduction attributed to LAPs (the ratio $\Delta\alpha_{MODIS,daily}^{LAPs}/\Delta\alpha_{in-situ,daily}^{LAPs}$ to be larger than 1).

5. Figure 1: In each panel, one parameter is varied while three are kept constant. What were the constant (i.e. default) values assumed in this figure?

R: We have added the assumed constant variabilities values in each panel in Figure 1:

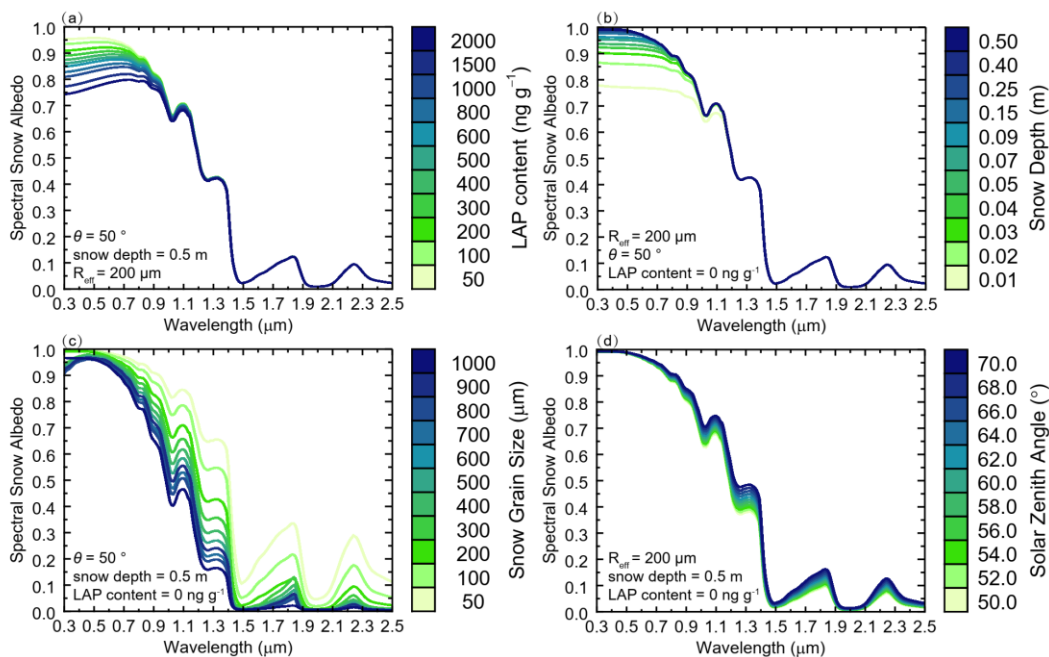


Figure 1. Variations in spectral snow albedo due to (a) LAP content (ng g^{-1}), (b) snow depth (m), (c) snow grain size (μm), and (d) solar zenith angle (deg.) while other three parameters are kept constant.

6. Table S1: In addition to MAE and RMSE, it would be useful to give the correlation coefficient between the corrected MODIS retrievals and the measurement-based albedo reductions.

R: We have added the correlation coefficient in Table S1:

	Northeastern China	Northwestern China	NA	Canadian Arctic	Greenland	Russian Arctic
MAE	0.064	0.016	0.014	0.0038	0.0014	0.011
RMSE	0.088	0.020	0.024	0.0075	0.0016	0.016
Correlation coefficient	0.13	-0.22	0.25	0.53	0.37	0.27

Technical and language corrections

- p. 3, line 8: Replace “radiances” with “radiation”. Also on p. 22, line 2.
R: Revised as suggestion.
- p. 9, line 18: “which briefly”. Something missing here?
R: Sorry for the grammar mistake and we have revised as “The equivalent BC has been defined by Doherty et al. (2010) which briefly as the amount of BC in the snow accounted for the wavelength-integrated total light absorption in the wavelengths of 300-750 nm by all particulate constituents.” in p. 9, line 18-21.
- p. 11, line 6: Replace “is performed” with “are assumed”.
R: Revised as suggestion.
- p. 15, line 4: Replace “competent” with “component”.
R: Revised as suggestion.
- p. 20, lines 15-17: Reformulation suggested, to improve clarity: “... and the results mainly represent winter for midlatitudes (because spring is mostly snow-free) and spring for the Arctic (because albedos cannot be derived during polar night)”.
R: Revised as suggestion.
- p. 21, line 2: “where is considerably higher”. Something is missing here. Should it be “where the emissions are considerably higher”?

R: Revised as suggestion.

7. p. 22, line 4: Remove “radiances”, or replace it with “radiative”, since “radiances flux” is not correct. “Radiance” refers to the intensity of radiation coming from a certain direction, and “radiative flux” (aka. “irradiance”) refers to the power radiated through a certain area, i.e., radiances integrated over a half-sphere.

R: Thank you for explaining and distinguishing the concept about “radiances”, “radiative”, “radiances flux” and “irradiance”. We have rechecked the errors and revised throughout the manuscript.

8. p. 27, line 16 (and Fig. 6 and Fig. S8): The terms “negative uncertainty” and “positive uncertainty” are not commonly used. Do you mean “the lower bound and the upper bound of the uncertainty range”?

R: Sorry for the non-standard terminology and revised as suggestion.

9. p. 27, lines 17-18: replace “by higher uncertainties” with “contributing more to the uncertainty”.

R: Revised as suggestion.

10. p. 28, lines 7-11: This is not expressed very clearly. What about: “MODIS has variably spaced and discrete spectral bands and thus cannot provide a continuous spectral measurement of reflectance. This results in a non-negligible uncertainty in retrieving the radiative forcing by LAPs in snow.”

R: Revised as suggestion.

11. p. 28, line 13: Should this be “a sample site located somewhere within the pixel”? (In-situ measurements are not necessarily taken at the midpoint of MODIS pixels).

R: Revised as suggestion.

12. p. 28, line 14: replace “true” with “representative”.

R: Revised as suggestion.

13. p. 30, line 2: replace “radiances” with “radiative fluxes”.

R: Revised as suggestion.

14. p. 31, line 13, and p. 35, lines 8-9: replace “Earth system modeling” with “CESM2”. (The performance of other Earth System Models might well differ from CESM2).

R: Revised as suggestion and we would like to add more modeling simulations to compare with our retrievals if these datasets were available in CMIP6 in the future.

15. Fig. 7: Thank you for including this figure! To improve its readability, please consider using a colour scale with other colours than just red and white.

R: Revised as suggestion:

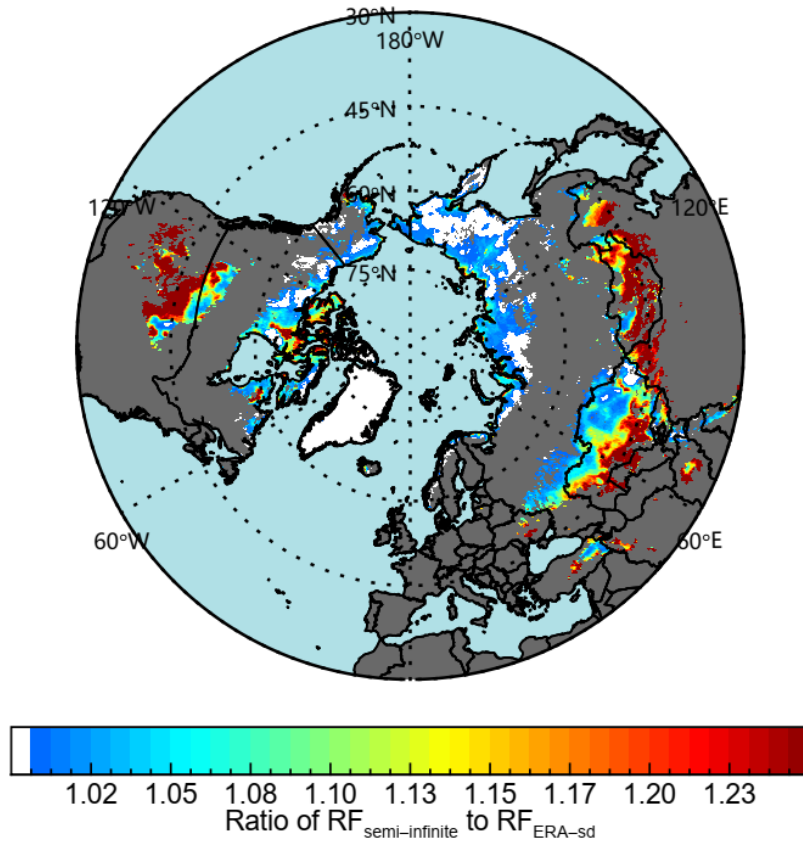


Figure 7. The spatial distribution of the ratio of retrieved radiative forcing using semi-infinite snow to radiative forcing using ERA-Interim snow depth.

16. Fig. 9 (upper panel): The geographic factor (G) seems not to appear at all in the colour bars. Is this an error, or is the contribution too small to be seen?

R: Actually, the geographic factor (G) makes too small contribution ($< 1\%$) to snow albedo reduction, both on regional and global scales.

17. Several of the figures in the Supplementary material (specifically, Figs. S1, S3, S4 and S6) would benefit from making the figure panels larger. Currently, a magnifying glass is required for reading the axis labels!

R: Revised as suggestion:

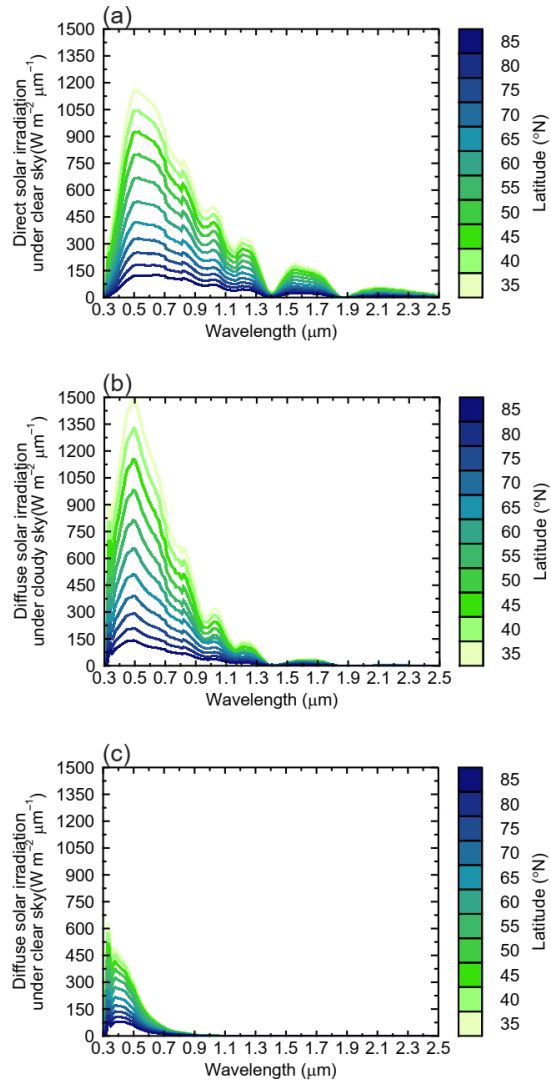


Figure S1. (a) Average December-May incident direct solar spectra for latitudes 35°–85°, derived from the SBDART model during clear-sky conditions. (b) Same as (a), but for diffuse solar irradiance. (c) Same as (a), but for cloudy-sky condition.

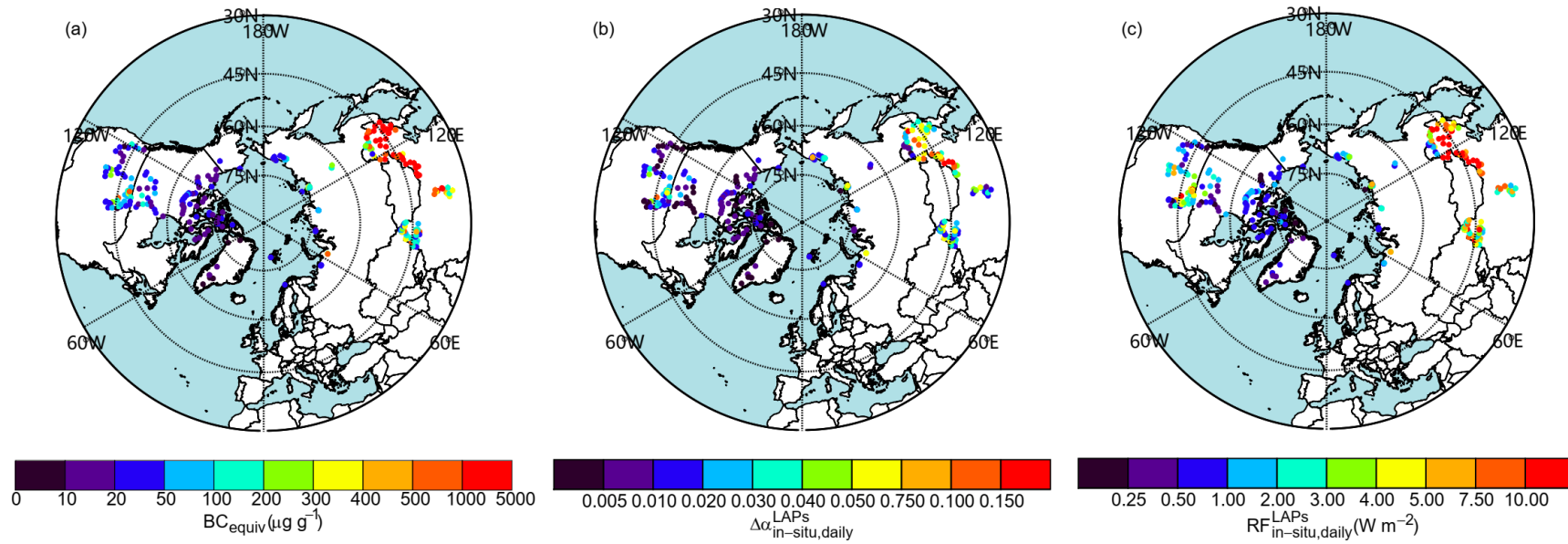


Figure S3. Spatial distribution of (a) in-situ measurements of BC_{equiv} and (b) the in-situ snow albedo reduction and (c) radiative forcing.

The snow albedo reduction and radiative forcing were calculated by SNICAR using measured BC_{equiv} .

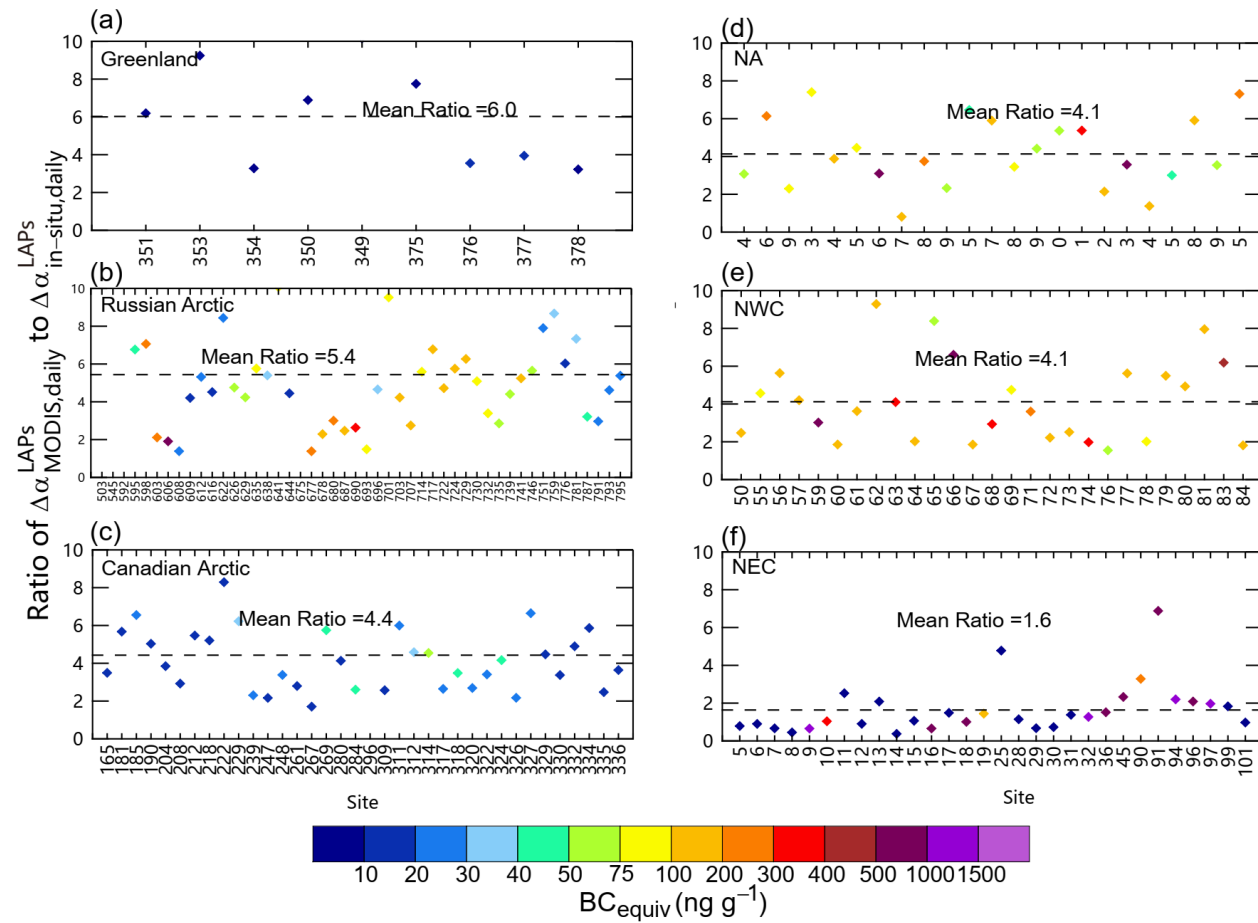


Figure S4. Ratio of $\Delta\alpha_{MODIS,ins}$ to $\Delta\alpha_{in-situ,ins}$. Panels (a)–(f) represent the snow samples collected in Greenland, Russian Arctic, Canadian Arctic, NA, NWC, and NEC, respectively.

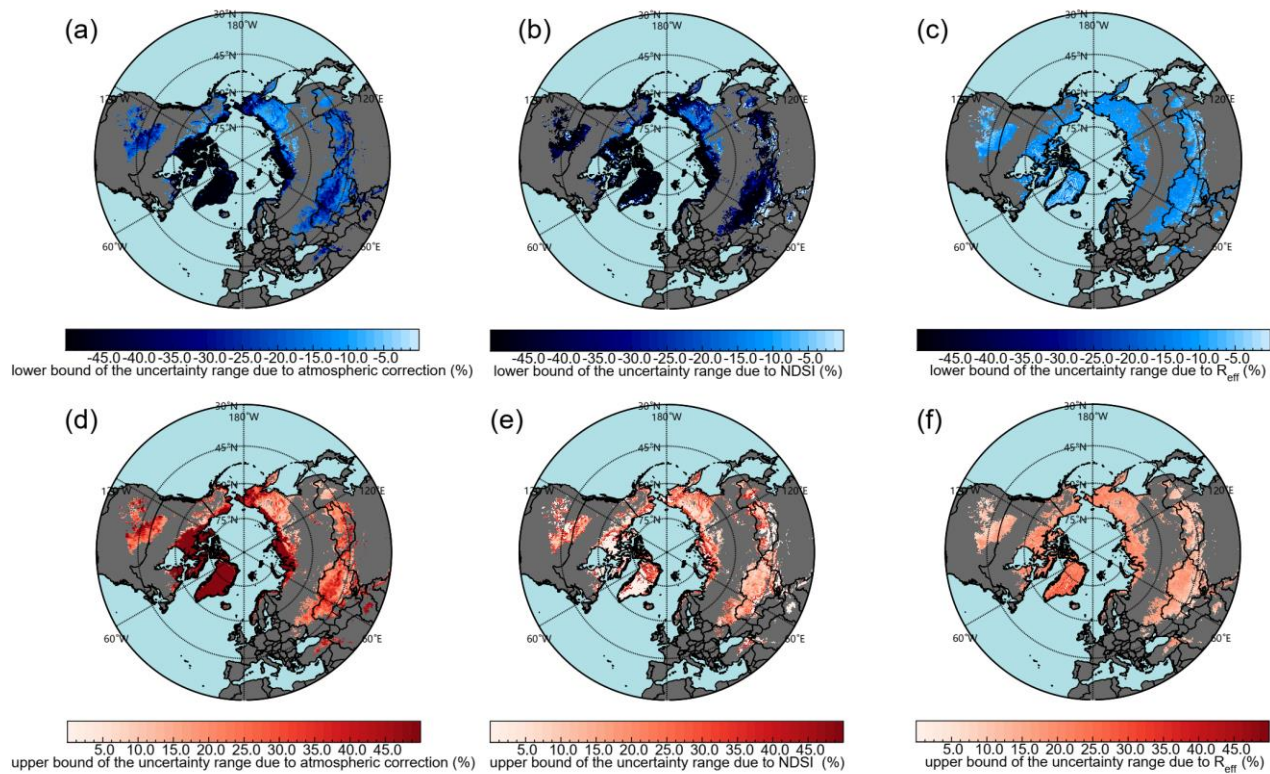


Figure S6. Spatial distributions of the lower bound of the uncertainty range due to (a) atmospheric correction, (b) snow cover fraction calculation and (c) snow grain size retrieval, respectively. (d)-(f) Same as (a)-(c), but for the upper bound of the uncertainty range.

References

Painter, T. H., Bryant, A. C., and Skiles, S. M.: Radiative forcing by light absorbing impurities in snow from MODIS surface reflectance data, *Geophysical Research Letters*, 39, n/a-n/a, 10.1029/2012gl052457, 2012

Manninen, T., Anttila, K., Jääskeläinen, E., Riihelä, A., Peltoniemi, J., Räisänen, P., Lahtinen, P., Siljamo, N., Thölix, L., Meinander, O., Kontu, A., Suokanerva, H., Pirazzini, R., Suomalainen, J., Hakala, T., Kaasalainen, S., Kaartinen, H., Kukko, A., Hautecoeur, O., and Roujean, J.-L.: Effect of small-scale snow surface roughness on snow albedo and reflectance, *The Cryosphere Discuss.*, <https://doi.org/10.5194/tc-2020-154>, in review, 2020.

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We are very grateful for the referee's constructive comments and suggestions throughout the peer-review process. Moreover, the referee's comments are quite significant that can help us to improve the paper quality substantially, especially in the uncertainty discussions and data availability. We have already rechecked the manuscript and made grammatical corrections and technical corrections.

1 Satellite-based radiative forcing by light-absorbing particles in snow across the
2 Northern Hemisphere

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10

1 **Abstract.** Snow is the most reflective natural surface on Earth and consequently plays
2 an important role in Earth's climate. Light-absorbing particles (LAPs) deposited on the
3 snow surface can effectively decrease snow albedo, resulting in positive radiative
4 forcing. In this study, we used remote sensing data from NASA's Moderate Resolution
5 Imaging Spectroradiometer (MODIS) and the Snow, Ice, and Aerosol Radiative
6 (SNICAR) model to quantify the reduction in snow albedo due to LAPs, before
7 validating and correcting the data against in-situ observations. We then incorporated
8 these corrected albedo-reduction data in the Santa Barbara DISORT Atmospheric
9 Radiative Transfer (SBDART) model to estimate Northern Hemisphere radiative
10 forcing except for midlatitude mountains in December-May for the period 2003–2018.
11 Our analysis reveals an average corrected reduction in snow albedo ($\Delta\alpha_{MODIS,corrected}^{LAPs}$)
12 of ~ 0.021 under all-sky condition, with daily radiative forcing ($RF_{MODIS,daily}^{LAPs}$) values
13 of $\sim 2.9 \text{ W m}^{-2}$, over land areas with complete or near-complete snow cover, with little
14 or no vegetation above the snow~~mapped snow-covered area~~ in Northern Hemisphere.
15 We also observed significant spatial variations in $\Delta\alpha_{MODIS,corrected}^{LAPs}$ and
16 $RF_{MODIS,daily}^{LAPs}$, with the lowest respective values (~ 0.016 and $\sim 2.6 \text{ W m}^{-2}$) occurring in
17 the Arctic and the highest (~ 0.11 and $\sim 12 \text{ W m}^{-2}$) in northeastern China. From MODIS
18 retrievals, we determined that the LAP content of snow accounts for 84% and 70% of
19 the spatial variability in albedo reduction and radiative forcing, respectively. We also
20 compared retrieved radiative forcing values with those of earlier studies, including
21 local-scale observations, remote-sensing retrievals, and model-based estimates.
22 Ultimately, estimates of radiative forcing based on satellite-retrieved data are shown to

1 represent true conditions on both regional and global scales.

2 **1. Introduction**

3 Seasonal snow cover affects 30% of Earth's land surface and exerts a cooling influence
4 on global climate through its direct interaction with the surface radiance budget
5 (Painter et al., 1998; Flanner et al., 2011). However, snow surface darkening due to
6 light-absorbing particles (LAPs) such as black carbon (BC), organic carbon (OC), dust,
7 and algae, can significantly alter the reflective properties of snow (Warren, 1982, 1984;
8 Hadley and Kirchstetter, 2012). When deposited on the snow surface, LAPs increase
9 the absorption of solar ~~radiation~~radiance (Painter et al., 2012a; Liou et al., 2014; Dang
10 et al., 2017), thereby reducing the snow albedo (Warren and Brandt, 2008; Kaspari et
11 al., 2014). As a result, radiative forcing of LAPs in snow (RFLS) plays a critical role in
12 snow-cover decline on both regional and global scales (Warren and Wiscombe, 1980),
13 perturbing the climate system and impacting hydrological cycles (Qian et al., 2011).

14 One of the primary LAPs, BC, is derived from the incomplete combustion of fossil
15 fuels and biomass (Bond et al., 2013; Dang et al., 2015) and is second only to CO₂ in
16 its contribution to climate forcing (Hansen and Nazarenko, 2004; Ramanathan and
17 Carmichael, 2008; Bond et al., 2013). Yet, despite considerable efforts to measure the
18 BC content of Northern Hemisphere snow and ice (Doherty et al., 2010, 2014; Huang
19 et al., 2011; Ye et al., 2012; Wang et al., 2013b, 2017), the inherent challenges presented
20 by a temporospatially variable snow cover mean our understanding of LAPs in snow is
21 far from complete. As a result, persistent uncertainties remain in regional and global-

1 scale RFLS estimates based on field measurements (Zhao et al., 2014).

2 Several previous investigations have utilized numerical models to estimate RFLS,
3 including that of Hansen and Nazarenko (2004), who concluded that BC in snow and
4 ice exerts a positive climate forcing throughout the Northern Hemisphere of $+0.3 \text{ W m}^{-2}$,
5 or explaining approximately one quarter of observed global warming. More recently,
6 Flanner et al. (2007) employed an aerosol/chemical-transport general-circulation model,
7 coupled with the Snow, Ice, and Aerosol Radiative (SNICAR) model (Flanner et al.,
8 2007; 2009), to estimate globally averaged radiative forcing values of $+0.054$ (range
9 $0.007\text{--}0.13$) and $+0.049$ ($0.007\text{--}0.12$) W m^{-2} for a strong (1998) and weak (2001) boreal
10 fire year, respectively. Using the Weather Research and Forecasting (WRF) model
11 (Skamarock et al., 2008) coupled with a chemistry component (Chem) (Grell et al.,
12 2005) and SNICAR modeling, Zhao et al. (2014) demonstrated that RFLS over northern
13 China in January–February 2010 was $\sim 10 \text{ W m}^{-2}$. However, despite their potentially
14 valuable contribution, climate models contain significant uncertainties in
15 representations of LAP emissions, transport, deposition, and post-depositional
16 processes that can propagate into simulations of LAP concentrations and their climate
17 forcing (Qian et al., 2015; Lee et al., 2016). Zhao et al. (2014) also confirmed that,
18 relative to observational data, modeled LAPs and radiative forcing estimates exhibit
19 biases that are difficult to explain and quantify. These shortcomings underscore the need
20 for a refined approach to estimating real-time RFLS that minimizes the mismatch
21 between field observations and model simulations.

1 In addition to modeling, remote sensing has been used to assess the physical
2 characteristics of snow cover (Nolin and Dozier, 1993, 2000; Painter et al., 2009, 2012a,
3 2013; Miller et al., 2016). Nolin and Dozier (2000), for example, retrieved grain-size
4 data from satellite-derived reflectance at near-infrared (NIR) wavelengths, following
5 the rationale that snow-grain size, in conjunction with solar zenith angle, dictates the
6 path-length of penetrating photons (Wiscombe and Warren, 1980) and thus influences
7 albedo in the NIR. Similarly, recent studies have attempted to employ satellite-derived
8 snow albedo at visible (VIS) wavelengths to retrieve RFLS data (Seidel et al., 2016; Pu
9 et al., 2019). Briefly, this retrieval method exploits the imaginary component of the
10 complex refractive index for ice (K_{ice}), which is very low at VIS wavelengths and
11 results in the extremely high VIS albedo for pure snow. In contrast, the imaginary
12 component of the complex refractive index for LAPs (K_{LAPs}) at VIS wavelengths is
13 orders of magnitude greater, resulting in the reduction in VIS snow albedo (Wiscombe
14 and Warren, 1980). Moreover, albedo variability at VIS wavelengths is dominated by
15 even minor concentrations of LAPs (Brandt et al., 2011; Painter et al., 2012b).

16 Painter et al. (2012a) employed surface-reflectance data provided by NASA's Moderate
17 Resolution Imaging Spectroradiometer (MODIS) for the Upper Colorado River Basin
18 and Hindu Kush-Himalaya (HKH) to make the first quantitative, remote-sensing-based
19 retrievals of instantaneous surface radiative forcing (RF) due to LAPs. Relative to the
20 Western Energy Balance of Snow (WEBS) network (Painter et al., 2007), that study
21 established that MODIS-derived radiative forcing exhibits a positive bias at lower RF

1 values and a slightly negative bias at higher values. A more recent study by Seidel et al.
2 (2016) used remote sensing to constrain instantaneous melt-season RFLS values of 20–
3 200 W m⁻² for the Sierra Nevada and Rocky Mountains, while Pu et al. (2019) reported
4 MODIS-derived values of 22–65 W m⁻² for northern China in January–February
5 (regional average ~45 W m⁻²). Acknowledging this demonstrated efficacy of remote
6 sensing retrievals for establishing RFLS on regional scales, we note this approach has
7 so far not captured spatial variability in RFLS on a global scale.

8 In this study, we employed MODIS data to determine the reduction in Northern
9 Hemisphere snow albedo due to LAPs. Retrievals were validated and corrected
10 according to ground-based snow observations, after which spatial variability in albedo
11 reduction and radiative forcing over mapped snow-covered area in Northern
12 Hemisphere were assessed quantitatively. Finally, we compared our satellite-derived
13 radiative forcing values with the modeling results of CESM2 (Eyring et al., 2016;
14 Danabasoglu et al., 2020). Despite the persistence of non-negligible uncertainties and
15 biases, our satellite-based retrievals constitute the first hemisphere-scale assessment of
16 RFLS and provide valuable information for improving climate model simulations.

17 **2. Data**

18 **2.1. Remote-sensing data**

19 To investigate the impact of LAPs on snow albedo, we utilized the following MODIS
20 data sets: surface albedo (MCD43C3; 0.05° × 0.05° resolution), snow cover
21 (MYD10C1; 0.05° × 0.05° resolution), land cover type (MCD12C1; 0.05° × 0.05°

1 resolution), and atmospheric parameters (MYD08_D3; $1^\circ \times 1^\circ$ resolution). Each data
2 set corresponds to December-May for the period 2003–2018 (<https://earthdata.nasa.gov>,
3 last access: 20 January 2019). MCD43C3 is the daily combined MODIS output derived
4 from both the Terra and Aqua satellites, and provides black-sky albedo (directional
5 hemispherical reflectance, DHF) and white-sky albedo (bi-hemispherical reflectance,
6 BHF) at local solar noon for bands 1–7 (band 1, 620–670 nm; band 2, 841–876 nm;
7 band 3, 459–479 nm; band 4, 545–565 nm; band 5, 1230–1250 nm; band 6, 1628–1652
8 nm; band 7, 2105–2155 nm), as well as values for quality control, local noon solar
9 zenith angle, and associated parameters. MCD43C3 observations are weighted to
10 estimate albedo on the 9th day of each 16-day period and have been corrected for the
11 influence of local slope and aspect, atmospheric gases, and aerosols.

12 Snow-cover data are provided daily by MYD10C1 as a report of the snow-cover
13 fraction (SCF), derived from the Normalized Difference Snow Index (NDSI).
14 MCD12C1 provides a spatially aggregated and reprojected land-cover type, which is
15 derived from the supervised classification of MODIS reflectance data, while MODIS
16 MYD08_D3 reports values of solar azimuth angle.

17 Average-daily solar radiances and cloud fraction were obtained from NASA’s Clouds
18 and the Earth’s Radiant Energy System (CERES: <https://ceres.larc.nasa.gov>, last access:
19 12 April 2019), part of the Earth Observing System comprising the Aqua, Terra, and S-
20 NPP satellites. CERES provides instantaneous measurements of solar radiances, which
21 are then converted to average-daily flux by angular dependence and empirical diurnal

1 albedo modeling as the satellite passes through the point of descent (Doelling et al.,
2 2013; Su et al., 2015; Loeb et al., 2018). We used the total downward shortwave flux
3 and cloud fraction at the surface, provided by the “CERES Single Scanner Footprint
4 1.08 (SSF1deg)” product, to estimate average-daily RFLS under all-sky conditions.
5 Shuttle Radar Topography Mission (SRTM) digital elevation data are provided by the
6 US Geological Survey (<https://www.usgs.gov/>, last access: 9 December 2018) to adjust
7 slope- and aspect-induced changes of surface solar irradiance in complex terrain. The
8 spatial resolution of SRTM data for the Northern Hemisphere is 30 m.

9 **2.2. Snow depth data**

10 Estimates of snow depth were obtained from the European Centre for Medium-Range
11 Weather Forecasts (ECMWF) Interim Re-Analysis (ERA-Interim)
12 (<https://www.ecmwf.int>, last access: 15 January 2019). ERA-Interim is a new
13 generation of reanalysis based on a 12-hourly and 4-dimensional variational data
14 assimilation (4D-Var) covering the period 1979–present. ERA-Interim performs better
15 in model physics frameworks, data quality control, and background error criteria than
16 previous versions (Berrisford et al., 2011; Brun et al., 2013). In this study, we used
17 snow-water equivalent (SWE) data for December–May covering the period 2003–2018.
18 These data were generated by forecast models and updated according to a Cressman
19 analysis of snow observations (Drusch et al., 2004; Dee et al., 2011). We note that the
20 previous occurrence of false snow-free patches, arising from application of Cressman
21 analysis in regions of sparse ground control, has been mitigated by ECMWF upgrades

1 (Dee et al., 2011). Finally, SWE is converted to snow depth by assuming that average
2 December-May snow density is $\sim 300 \text{ kg m}^{-3}$, consistent with snow-depth estimates by
3 the Canadian Meteorological Centre (CMC) (Sturm et al., 1995; Brown and Mote,
4 2009).

5 **2.3. In-situ measurements of LAPs in snow**

6 To correct the satellite retrievals, we collected a comprehensive set of in-situ
7 measurements of BC concentrations from the field campaigns in the Arctic in spring of
8 2005-2009 (Doherty et al., 2010), North America in January-March of 2013 (Doherty
9 et al., 2014), Northern China in January-February of 2010, 2012 and 2014 (Ye et al.,
10 2012; Wang et al., 2013; Wang et al., 2017). The BC concentrations are measured by
11 the two-sphere integrating-sandwich (TSI) spectrophotometer in the Arctic, North
12 America, and Northern China (Grenfell et al., 2011; Wang et al., 2020). Briefly, TSI
13 produces a diffuse radiation field when the white light illumination is transmitted into
14 an integrating sphere; then the diffuse radiation passes through the filter and is detected
15 by a spectrometer. The TSI technique acquires the light attenuation spectrum due to the
16 LAPs loaded on the sample filter (Grenfell et al., 2011). Then, the light attenuation
17 spectrum of the sample filter is transformed into an equivalent BC mass (unit: g cm^{-2})
18 loading by comparing against the standard filters. The equivalent BC has been defined
19 by Doherty et al. (2010) which briefly as the amount of BC in the snow ~~to~~-accounted
20 for the wavelength-integrated total light absorption in the wavelengths of 300-750 nm
21 by all particulate constituents. In this study, we used BC_{equiv} for all LAPs to calculate

1 the in-situ snow albedo reduction and radiative forcing (Fig. S3).

2 **2.4. Climate model simulations**

3 We compared our remotely sensed retrievals of daily-average RFLS for the 2003–2014
4 study period with simulated results derived from CESM2 (<https://esgf-node.llnl.gov/>,
5 last access: 15 July 2019). In this study, we employed simulations of snow BC
6 concentrations derived from the CESM2 historical experiments, in conjunction with
7 ERA-Interim SWE, MODIS-retrieved snow grain-size, and CERES total downward
8 shortwave flux data under all-sky condition, to model daily-average RFLS for the study
9 period. Simulations were performed using the Snow, Ice, and Aerosol Radiative
10 (SNICAR) and Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART)
11 models, and the model output was compared with satellite-based retrievals.

12 **3. Methods**

13 **3.1. Radiative transfer model**

14 In this study, we used the Santa Barbara DISORT Atmospheric Radiative Transfer
15 (SBDART) model to calculate spectral surface solar irradiance. Constituting one of the
16 most widely applied models for calculating the atmospheric radiative transfer at Earth's
17 surface, under both clear- and cloudy-sky conditions (Ricchiazzi et al., 1998), SBDART
18 combines a low-resolution atmospheric transmission model, Discrete Ordinate
19 Radiative Transfer (DISORT) module, and Mie scattering output for the scattering of
20 light by ice crystals and water droplets (Stamnes et al., 1988; Fu et al., 2017). Radiative
21 transfer equations for a vertically inhomogeneous, non-isothermal, plane-parallel

1 atmosphere are integrated numerically using the DISORT module. SBDART comprises
2 multiple standard atmospheric profiles, cloud models, basic surface types, as well as
3 vertical distribution models for aerosols and gas absorption, and enables users to specify
4 these input parameters in real values. In our study, the subarctic and midlatitude winter
5 standard atmospheric condition ~~is performed~~are assumed as well as the tropospheric
6 and stratospheric background aerosols are archived in SBDART (Tanre, D. et al., 1990).
7 According to Dang et al. (2017), the cloud optical depth in high-latitude and mid-
8 latitude was assumed as 11 and 20 under cloudy-sky condition, respectively. The
9 spectral irradiance from SBDART is only used for integrating the spectral MODIS
10 albedo to achieve broadband albedo, thus the uncertainty of solar irradiance from the
11 assumed atmospheric properties has limited influence on the retrieval of radiative
12 forcing (see Section 3.2). Average incident direct and diffuse solar spectra for
13 December-May under clear/cloudy sky are shown in Fig. S1.

14 The Snow, Ice, and Aerosol Radiative (SNICAR) model is a two-stream multiple
15 scattering radiative transfer model (Flanner et al., 2007, 2009) that has been used widely
16 both to simulate the albedo, transmission, and vertical absorptivity of LAP-
17 contaminated snowpack and to estimate RFLS (Painter et al., 2012a; Bryan et al., 2013;
18 Miller et al., 2016). SNICAR employs the theory proposed by Wiscombe and Warren
19 (1980) and Toon et al. (1989). Specifically, snow is considered to be composed of
20 aggregated ice grains with optical effective radii (R_{eff}) of 50–1500 μm , lognormal
21 distribution, and spherical grain shape. SNICAR also accounts for the incident radiation

1 at the surface and its spectral distribution, solar zenith angle, snow depth and density,
2 snow layer number, and the type and concentration of LAPs in the snowpack. The
3 model's ability to provide realistic simulations of snow albedo has been verified by
4 several previous studies (Hadley and Kirchstetter, 2012; Meinander et al., 2013; Zhong
5 et al., 2017; Wang et al., 2017).

6 **3.2. Retrieval of quantitative snow properties from remote sensing**

7 The variability of spectral snow albedo depends on the LAP content, grain size, grain
8 shape, and depth of the snowpack, in addition to solar zenith angle. As shown in Fig.
9 1a, the deposition of BC (as representative of LAPs generally) serves to decrease the
10 albedo of snow significantly, particularly in the ultraviolet (UV) and VIS wavelengths,
11 which account for approximately half of all direct solar irradiance and the majority of
12 diffuse solar irradiance (Fig. S1). In contrast, the impact of BC on albedo is
13 considerably smaller in NIR wavelengths and can be negligible at $> \sim 1150$ nm. Snow
14 depth plays a similar role to LAP content and primarily affects albedo in UV and VIS
15 wavelengths (Fig. 1b).

16 Although snow albedo decreases with snow depth, previous studies have tended to
17 assume a semi-infinite snowpack for which albedo is independent of depth. As a
18 consequence, the role of LAPs in albedo reduction has been overestimated for those
19 areas where the snowpack is thin (Warren, 2013). In this study, we incorporated ERA-
20 Interim SWE data in our SNICAR model simulations to correct for the snow-depth
21 overestimation effect. In contrast, snow grain-size and solar zenith angle influence the

1 snow albedo chiefly in NIR wavelengths (Fig. 1c, d). Specifically, albedo tends to
 2 decrease with increasing snow grain-size and declining solar zenith angle. In this study,
 3 we derived quantitative snow parameters (grain size, albedo reduction, and RFLS) from
 4 MODIS data in conjunction with the SNICAR and SBDART models. The specific
 5 workflow for retrieving RFLS from satellite data is shown in Fig. 2.

6 **3.2.1. Retrieval of blue-sky albedo**

7 MCD43 provides black-sky and white-sky albedo, which are defined as albedo in the
 8 absence of diffuse and direct component of solar irradiance. Accordingly, the actual
 9 spectral albedo for a land surface at wavelength λ (also called blue-sky albedo:
 10 $\alpha_{MODIS,\lambda}^{blue-clear}$) under clear-sky condition can be calculated as follows:

$$11 \quad \alpha_{MODIS,\lambda}^{blue-clear} = f_{dif,\lambda}^{clear} \cdot \alpha_{MODIS,\lambda}^{white-sky} + (1 - f_{dif,\lambda}^{clear}) \cdot \alpha_{MODIS,\lambda}^{black-sky} \quad (1)$$

12 where $\alpha_{MODIS,\lambda}^{white-sky}$ and $\alpha_{MODIS,\lambda}^{black-sky}$ are MODIS-derived values for white-sky and
 13 black-sky albedo, respectively, and $f_{dif,\lambda}^{clear}$ is the ratio of diffuse irradiance to the total
 14 solar irradiance under clear-sky (Lewis and Barnsley, 1994). The latter is calculated as
 15 follows:

$$16 \quad f_{dif,\lambda}^{clear} = \frac{E_{dif}^{clear}(\lambda; \varphi)}{E_{dif}^{clear}(\lambda; \varphi) + E_{dir}^{clear}(\lambda; \varphi) \cdot \cos\beta} \quad (2)$$

17 where φ is latitude, and $E_{dif}^{clear}(\lambda; \varphi)$ denote the diffuse spectral irradiance on a
 18 horizontal surface and $E_{dir}^{clear}(\lambda; \varphi)$ denote the direct spectral irradiance on a surface
 19 perpendicular to the sun~~diffuse and direct spectral solar irradiance, respectively~~, derived
 20 from the SBDART model under clear-sky condition. β represents local solar zenith

1 angle, which is obtained using the topographic correction method (Teillet et al., 1982;
 2 Negi and Kokhanovsky, 2011):

$$3 \quad \cos \beta = \cos \theta_0 \cos \theta_T + \sin \theta_0 \sin \theta_T \cos(\phi_0 - \phi_T) \quad (3)$$

4 for which θ_0 represents the solar zenith angle for a horizontal surface, ϕ_0 is the solar
 5 azimuth angle, and θ_T and ϕ_T denote slope inclination and aspect, respectively.

6 Similarly, we can derive the blue-sky albedo for cloudy-sky condition ($\alpha_{MODIS,\lambda}^{blue-cloudy}$).

7 Then, we used cloud fraction (f_{cloud}) from CERES to weight clear-sky albedo and
 8 cloudy-sky albedo to obtain actual all-sky albedo ($\alpha_{MODIS,\lambda}^{all}$):

$$9 \quad \alpha_{MODIS,\lambda}^{all} = f_{cloud} \cdot \alpha_{MODIS,\lambda}^{blue-cloudy} + (1 - f_{cloud}) \cdot \alpha_{MODIS,\lambda}^{blue-clear} \quad (4)$$

10 **3.2.2. Retrieval of snow cover and albedo values**

11 As shown in Fig. 2, the snow-covered area is mapped according to the actual all-sky
 12 albedo ($\alpha_{MODIS,\lambda}^{all}$) in band 4 (band center ~555 nm) and the Normalized Difference
 13 Snow Index (NDSI), both of which are required to exceed 0.6 (Negi and Kokhanovsky,
 14 2011). According to the MODIS Snow Products Collection 6 User Guide
 15 (<http://nsidc.org/data>), the Fractional Snow Cover (FSC) can be calculated as follows:

$$16 \quad FSC = -0.01 + 1.45 \cdot NDSI \quad (5)$$

17 Accordingly, the identified snow-covered area (ISCA) has an FSC value of >86% but
 18 not always 100%. Therefore, the MODIS-derived albedo for a particular ISCA is a
 19 combination of values representing both snow and the snow-free underlying surface.

20 Following Pu et al. (2019), the snow albedo ($\alpha_{snow,\lambda}^{all}$) can be distinguished from the

1 mixed albedo by the equation:

$$2 \quad \alpha_{MODIS,\lambda}^{all} = \frac{E_{all-sky,\lambda} \cdot FSC \cdot \alpha_{snow,\lambda}^{all} + E_{all-sky,\lambda} \cdot (1 - FSC) \cdot \alpha_{underlying,\lambda}}{E_{all-sky,\lambda}}$$

$$3 \quad = FSC \cdot \alpha_{snow,\lambda}^{all} + (1 - FSC) \cdot \alpha_{underlying,\lambda} \quad (6)$$

$$4 \quad \alpha_{snow,\lambda}^{all} = \frac{\alpha_{MODIS,\lambda}^{all} - (1 - FSC) \cdot \alpha_{underlying,\lambda}}{FSC} \quad (7)$$

5 where $E_{all-sky,\lambda}$ is total solar irradiance under all-sky condition, a linear combination
6 of direct/diffuse competent component of solar irradiance under clear-sky and cloudy-
7 sky using similar strategy via Eq. (1)-(4). $\alpha_{underlying,\lambda}$ represents the albedo of the
8 underlying surface and was obtained from Siegmund and Menz (2005). As depicted in
9 Fig. 3b, vegetation and bare soil are the main types of underlying surface in the ISCA.

10 **3.2.3. Retrieval of snow grain size**

11 The snow optical-equivalent grain size (R_{eff}) is retrieved by fitting SNICAR-simulated
12 snow albedo to MODIS-derived snow albedo at 1240 nm (the central wavelength of
13 MODIS band 5), following the protocol of Nolin and Dozier (2000). This retrieval
14 method is not influenced by liquid water and water vapor and has been employed
15 widely in previous studies (e.g., Painter et al., 2013; Seidel et al, 2016). Both Nolin and
16 Dozier (2000) and Pu et al. (2019) reported that the retrieved R_{eff} compares favorably
17 with ground-based measurements of snow grain size. In this study, we chose to exclude
18 the ISCA, where MODIS-derived snow albedo at 1240 nm is <0.3 , to avoid
19 misrepresenting R_{eff} (Tedesco et al., 2007).

20 **3.2.4. Retrieval of snow albedo reduction and RFLS**

1 The spectrally integrated reduction in snow albedo due to LAPs ($\Delta\alpha_{MODIS,noon}^{LAPs}$) is
 2 estimated for local-noon and all-sky conditions, using solar irradiance and the
 3 difference between MODIS-derived spectral snow albedo ($\alpha_{snow,\lambda}^{all}$) and simulated pure
 4 snow albedo ($\alpha_{snow,\lambda}^{mdl}$). Because MODIS provides only four VIS bands, we fitted snow
 5 albedo data obtained via MODIS to a continuous 300–2500 nm spectrum ($\alpha_{snow,\lambda}^{MODIS}$, with
 6 a 10 nm interval) following the method provided by Pu et al. (2019). Thereafter, the
 7 broadband albedo reduction due to LAPs retrieved from MODIS ($\Delta\alpha_{MODIS,noon}^{LAPs}$) can
 8 be calculated as follows:

$$9 \quad \Delta\alpha_{MODIS,noon}^{LAPs} = \frac{\sum_{\lambda=300nm}^{\lambda=2500nm} (\alpha_{snow,\lambda}^{mdl} - \alpha_{snow,\lambda}^{MODIS}) \cdot E_{all-sky,\lambda} \cdot \Delta\lambda}{\sum_{\lambda=300nm}^{\lambda=2500nm} E_{all-sky,\lambda} \cdot \Delta\lambda} \quad (8)$$

10 where $\alpha_{snow,\lambda}^{mdl}$ is the pure snow albedo simulated by SNICAR using MODIS-derived
 11 R_{eff} and ERA-Interim snow depth data, $\alpha_{snow,\lambda}^{MODIS}$ is the continuous snow albedo
 12 derived from MODIS retrievals, and $\Delta\lambda$ is 10 nm.

13 Following Miller et al. (2016), we assumed that the properties for snow and LAPs
 14 remain invariable throughout the day. Based on calculated $\alpha_{snow,\lambda}^{mdl}$ and $\alpha_{snow,\lambda}^{MODIS}$ at
 15 noon, the diurnal variation of pure and polluted snow albedo can be simulated by
 16 SNICAR from sunrise to sunset. Then, daily-average snow albedo reduction
 17 ($\Delta\alpha_{MODIS,daily}^{LAPs}$) can be derived by integrating the diurnal snow albedo reduction, which
 18 is weighted by simultaneous solar irradiance from SBDART. Similarly, we used
 19 measurements of LAPs in contaminated snow to calculate the
 20 in-situ reduction in snow albedo ($\Delta\alpha_{in-situ,daily}^{LAPs}$). To derive a correction factor for

1 MODIS retrievals, we applied a similar validation strategy to that of Zhu et al. (2017):

$$2 \quad c = \frac{1}{n} \sum_{i=1}^n \left(\frac{\Delta\alpha_{MODIS,daily}^{LAPs}}{\Delta\alpha_{in-situ,daily}^{LAPs}} \right) \quad (9)$$

3 where c is the correction factor for $\Delta\alpha_{MODIS,daily}^{LAPs}$ and n is the number of the
 4 respective in-situ measurements. Accordingly, the corrected albedo reduction
 5 ($\Delta\alpha_{MODIS,corrected}^{LAPs}$) is calculated as follows:

$$6 \quad \Delta\alpha_{MODIS,corrected}^{LAPs} = \frac{1}{c} \cdot \Delta\alpha_{MODIS,daily}^{LAPs} \quad (10)$$

7 The daily-average, spectrally integrated RFLS ($RF_{MODIS,daily}^{LAPs}$) is calculated for all-sky
 8 conditions as follows:

$$9 \quad RF_{MODIS,daily}^{LAPs} = \Delta\alpha_{MODIS,corrected}^{LAPs} \cdot SW_{all-sky} \quad (11)$$

10 where $SW_{all-sky}$ represent the average-daily total downward shortwave fluxes,
 11 obtained from CERES under all-sky conditions.

12 **3.2.5. Attribution of spatial variability in snow albedo reductions and radiative** 13 **forcing**

14 As demonstrated above, reductions in snow albedo and RFLS are dependent primarily
 15 on LAP content, R_{eff} , snow depth (SD), solar zenith angle, surface topography, and
 16 solar irradiance, the latter three of which can be categorized as the geographic factor
 17 (G). We used an impurity index (I_{LAPs}) to represent the LAP content of the snowpack
 18 (Di Mauro et al., 2015; Pu et al., 2019), following the equation:

$$19 \quad I_{LAPs} = \frac{\ln(\alpha_{snow,band4}^{all})}{\ln(\alpha_{snow,band5}^{all})} \quad (12)$$

1 where $\alpha_{snow,band4}^{all}$ and $\alpha_{snow,band5}^{all}$ are the MODIS-derived snow albedo values for
 2 bands 4 and 5, respectively. We then calculated $\Delta\alpha_{MODIS,corrected}^{LAPs}$ as follows:

$$3 \quad \Delta\alpha_{MODIS,corrected}^{LAPs} = f(I_{LAPs}, R_{eff}, SD, G) \quad (13)$$

4 The spatial variability in snow albedo reduction due to I_{LAPs} can be expressed as

$$5 \quad \Delta\alpha_{MODIS,corrected}^{LAPs}(I_{LAPs}) = f(I_{LAPs}, \overline{R_{eff}}, \overline{SD}, \overline{G}) \quad (14)$$

6 where $\overline{R_{eff}}, \overline{SD}, \overline{G}$ indicate spatial-mean values of $R_{eff}, SD,$ and $G,$ with \overline{G}
 7 requiring spatially constant values for the solar zenith angle, surface topography, and
 8 solar irradiance parameters. The following three equations were applied in a similar
 9 manner:

$$10 \quad \Delta\alpha_{MODIS,corrected}^{LAPs}(R_{eff}) = f(\overline{I_{LAPs}}, R_{eff}, \overline{SD}, \overline{G}) \quad (15)$$

$$11 \quad \Delta\alpha_{MODIS,corrected}^{LAPs}(SD) = f(\overline{I_{LAPs}}, \overline{R_{eff}}, SD, \overline{G}) \quad (16)$$

$$12 \quad \Delta\alpha_{MODIS,corrected}^{LAPs}(G) = f(\overline{I_{LAPs}}, \overline{R_{eff}}, \overline{SD}, G) \quad (17)$$

13 We then fitted $\Delta\alpha_{MODIS,corrected}^{LAPs}$ through multiple linear regression:

$$14 \quad \Delta\alpha_{MODIS}^{LAPs,fit} = a \cdot \Delta\alpha_{MODIS,corrected}^{LAPs}(I_{LAPs}) + b\Delta\alpha_{MODIS,corrected}^{LAPs}(R_{eff}) + c \cdot$$

$$15 \quad \Delta\alpha_{MODIS,corrected}^{LAPs}(SD) + d \cdot \Delta\alpha_{MODIS,corrected}^{LAPs}(G) \quad (18)$$

16 where $\Delta\alpha_{MODIS}^{LAPs,fit}$ is the fitted snow albedo reduction and a, b, c, and d denote the
 17 regression coefficients. Figure S3a illustrates how $\Delta\alpha_{MODIS}^{LAPs,fit}$ can explain 99% of the
 18 variance in $\Delta\alpha_{MODIS,corrected}^{LAPs}$. Therefore, the attribution of spatial variance in
 19 $\Delta\alpha_{MODIS,corrected}^{LAPs}$ can be replaced with $\Delta\alpha_{MODIS}^{LAPs,fit}$, enabling Eq. (18) to be written as

1 follows:

$$\begin{aligned}
2 \quad & \Delta\alpha_{MODIS}^{LAPs,fit} - \overline{\Delta\alpha_{MODIS}^{LAPs,fit}} = a \cdot (\Delta\alpha_{MODIS,corrected}^{LAPs}(I_{LAPs}) - \\
3 \quad & \overline{\Delta\alpha_{MODIS,corrected}^{LAPs}(I_{LAPs})}) + b \cdot (\Delta\alpha_{MODIS,corrected}^{LAPs}(R_{eff}) - \\
4 \quad & \overline{\Delta\alpha_{MODIS,corrected}^{LAPs}(R_{eff})}) + c \cdot (\Delta\alpha_{MODIS,corrected}^{LAPs}(SD) - \\
5 \quad & \overline{\Delta\alpha_{MODIS,corrected}^{LAPs}(SD)}) + d \cdot (\Delta\alpha_{MODIS,corrected}^{LAPs}(G) - \overline{\Delta\alpha_{MODIS,corrected}^{LAPs}(G)}) \quad (19)
\end{aligned}$$

6 where $\Delta\alpha_{MODIS}^{LAPs,fit} - \overline{\Delta\alpha_{MODIS}^{LAPs,fit}}$ is the snow albedo reduction anomaly
7 $(\Delta\alpha_{MODIS,anomaly}^{LAPs,fit})$. Then, Eq. (19) can be written as

$$\begin{aligned}
8 \quad & \Delta\alpha_{MODIS,anomaly}^{LAPs,fit} = a \cdot \Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(I_{LAPs}) + b \cdot \\
9 \quad & \Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(R_{eff}) + c \cdot \Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(SD) + d \cdot \\
10 \quad & \Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(G). \quad (20)
\end{aligned}$$

11 According to Huang and Yi (1991) and Pu et al. (2019), the fractional contribution of
12 LAP content to the variability in snow albedo reduction ($R_{\Delta\alpha}^{LAPs}$) can be calculated as:

$$13 \quad R_{\Delta\alpha}^{LAPs} = \frac{1}{m} \sum_{j=1}^m \frac{(\Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(I_{LAPs})_j)^2}{K_j} \quad (21)$$

$$\begin{aligned}
14 \quad & K_j = (a \cdot \Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(I_{LAPs})_j)^2 + (b \cdot \\
15 \quad & \Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(R_{eff})_j)^2 + (c \cdot \Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(SD)_j)^2 + \\
16 \quad & (d \cdot \Delta\alpha_{MODIS,corrected,anomaly}^{LAPs}(G)_j)^2 \quad (22)
\end{aligned}$$

17 where m denotes the length of the data set. Values for $R_{\Delta\alpha}^{R_{eff}}$, $R_{\Delta\alpha}^{SD}$, and $R_{\Delta\alpha}^G$ can be
18 derived in the same way. Similarly, we can obtain the fractional contribution for daily
19 radiative forcing (R_{RF}^{LAPs} , $R_{RF}^{R_{eff}}$, R_{RF}^{SD} , and R_{RF}^G).

1 4. Results

2 4.1. Study area

3 Figure 3a depicts the ISCA employed in this study. Most are located in Eurasia, North
4 America and the Arctic, which are dominated by grassland, shrublands and bare-soil
5 surfaces (Fig. 3b). Several mid–high-latitude regions that typically support a deep
6 snowpack, including southern Russia, western Europe, and eastern US, are not
7 identified by MODIS as ISCA due to the broad distributions of forest in those areas
8 (Fig. 3b). This pattern is supported by Bond et al. (2006), who demonstrated that, under
9 such vegetated conditions, LAPs in snow exert a relatively minor influence on radiative
10 forcing. On the other hand, the snowpack over midlatitude mountains at such a coarse
11 resolution ($0.05^\circ \times 0.05^\circ$) is too low to identify. In addition, midlatitude mountains are
12 characterized as complex terrain, which will lead to high biases in radiative forcing
13 retrieval at the coarse resolution in spite of topographic correction. Therefore, we didn't
14 report the results over midlatitude mountains in this study.

15 As illustrated in Fig. 3a, ISCA can be separated into four general regions according to
16 geographical distribution and pollution conditions (Fig. S2a, b): northeastern China
17 (NEC), Eurasia (EUA), North America (NA), and the Arctic. The following analysis of
18 snow albedo reduction and RFLS only concerns ISCA ~~and the periods of the results are~~
19 ~~mainly in winter for midlatitudes due to snow melting and in spring for the Arctic due~~
20 ~~to polar night, and the results mainly represent winter for midlatitudes (because spring~~
21 ~~is mostly snow-free) and spring for the Arctic (because albedos cannot be derived~~

1 during polar night).

2 **4.2. Global characteristics**

3 Previous studies have highlighted the dominant role of BC in light absorption by snow
4 (Wang et al., 2013b; Dang et al., 2017). The spatial distribution of BC emissions density
5 for the Northern Hemisphere in December-May is shown in Fig. S2a. Emissions density
6 exhibits a strong spatial inhomogeneity, ranging from $<10^{-1}$ to $>10^4$ g km⁻² month⁻¹
7 over ISCA. The highest values occur in NEC, where the emissions are considerably
8 higher~~where is considerably higher~~ than EUA and NA, and the lowest values occur in
9 the Arctic. The wet and dry deposition of BC constitute the primary mechanisms for
10 BC accumulation in snow. As shown in Fig. S2b, the distribution of BC deposition (i.e.,
11 the sum of dry and wet deposition) is similar to BC emissions density, with the highest
12 and lowest regional averages corresponding to NEC and the Arctic, respectively.
13 Together, these data indicate that the NEC snowpack is heavily polluted, and thus snow
14 albedo reduction is likely to be highest, while the Arctic snowpack is the least
15 contaminated.

16 In addition to LAP content, the physical properties of the snowpack, such as depth and
17 grain size, also impact snow albedo (Fig. 1). As depicted in Fig. 4a, the average
18 snowpack in EUA (0.15 m thick) is thinner than in both NA (0.24 m) and NEC (0.19
19 m), implying a greater impact of snow depth on snow albedo and radiative forcing in
20 EUA. The greatest snow depths occur in the Arctic (>1 m) and can be considered semi-
21 infinite, meaning that the impact of depth on albedo and radiative forcing is negligible.

1 Figure 4b shows the spatial distribution of MODIS-derived snow grain radius (R_{eff}).
2 In contrast to BC emissions density, BC deposition, and snow depth, R_{eff} exhibits
3 minor spatial variability, with regional average values for NEC, EUA, NA, and the
4 Arctic of 237 μm , 227 μm , 237 μm , and 215 μm , respectively. These values align with
5 the findings of several previous studies (Painter et al., 2013; Seidel et al, 2016; Pu et
6 al., 2019) and imply that the contribution of R_{eff} to spatial variability in snow albedo
7 reduction and radiative forcing is negligible.

8 According to Eq. (11), local solar ~~radiation~~ radiance is an important factor for
9 determining RFLS. Figure 4c depicts the December-May averaged total downward
10 surface shortwave flux under all-sky conditions. Average solar ~~radiance~~ radiative flux
11 values for EUA and NA are comparable to one another but high relative to NEC, which
12 lies at a generally higher latitude ($>40^\circ$). The lowest values occur in the Arctic due to
13 that region's extreme latitude. The Arctic goes through the polar night during winter,
14 so that the radiative effect of LAPs in the Arctic mainly appears in spring. Figure S2d
15 shows the March-May averaged downward surface shortwave flux. As can be seen that
16 the values in the Arctic in March-May are higher than those in midlatitudes in
17 December-February (Figure S2c). We note that snow albedo reduction and radiative
18 forcing are only calculated over the period when snow-covered area was mapped, which
19 implies that the RFLS will be higher in the Arctic than midlatitudes for the same snow
20 albedo reduction.

21 4.3. Corrections based on in-situ observations

1 Albedo reduction calculated using in-situ observed LAPs ($\Delta\alpha_{in-situ,daily}^{LAPs}$) were used to
2 quantitatively correct MODIS retrievals through comparison with MODIS-retrieved
3 snow albedo reduction ($\Delta\alpha_{MODIS,daily}^{LAPs}$). Figure S4 displays scatterplots of the ratios of
4 $\Delta\alpha_{MODIS,daily}^{LAPs}$ to $\Delta\alpha_{in-situ,daily}^{LAPs}$ ($r_{in-situ}^{MODIS}$) for each sampling sites (Ye et al., 2012;
5 Wang et al., 2013b, 2017; Doherty et al., 2010; 2014). Briefly, for NA, EUA, and the
6 Arctic where the snowpack is relatively clean, the values for $r_{in-situ}^{MODIS}$ mostly range
7 between 2 and 10. In contrast, the heavily polluted snowpack in NEC returns $r_{in-situ}^{MODIS}$
8 values ranging from 0.5 to 2.5, indicating a negative correlation between the biases of
9 $\Delta\alpha_{MODIS,daily}^{LAPs}$ and snow contamination, and thus supporting the findings of previous
10 studies (Painter et al., 2012a; Pu et al., 2019). To improve the quality of MODIS
11 retrievals, we developed the correction factors for different regions. According to Eq.
12 (10), the correction factors for NEC, EUA, NA, Canadian Arctic, Russian Arctic and
13 Greenland are 1.6, 4.1, 4.1, 4.4, 5.4 and 6.0, respectively. Hereafter, our analyses are
14 based on the corrected MODIS retrievals.

15 Figure 5 compares the corrected MODIS retrievals to measurement-based results, and
16 the mean absolute error (MAE) and root mean square error (RMSE) of
17 $\Delta\alpha_{MODIS,corrected}^{LAPs}$ relative to $\Delta\alpha_{in-situ,daily}^{LAPs}$ are given in Table S1. Together, these
18 results imply that the corrected MODIS retrievals are plausible. Nevertheless, we note
19 that the correction used in this study is spatially rough due to the low density of in-situ
20 measurements, thus that both the uncertainty and bias are non-negligible. To address
21 this issue, we presented further discussion about the accuracy of radiative forcing

1 retrievals (see Sect. 4.5). We also conducted a comprehensive series of comparisons
2 between the MODIS-derived retrievals and values provided via surface measurements,
3 model simulations, and remote sensing (see Sect. 5). We concluded that further field-
4 based measurements of snow albedo are required to improve the quality of satellite
5 retrievals.

6 **4.4. Spatial distributions of snow albedo reduction and radiative forcing**

7 Figure 6a shows the spatial distributions of MODIS-based albedo reduction and daily
8 radiative forcing, and statistics are shown in Figure 6b and Table 1. On average,
9 $\Delta\alpha_{MODIS,corrected}^{LAPs}$, and $RF_{MODIS,daily}^{LAPs}$ provide respective values of 0.021 and 2.9 W
10 m^{-2} for Northern Hemisphere ISCA. The highest $\Delta\alpha_{MODIS,corrected}^{LAPs}$ occurs in NEC,
11 where the regional average of ~ 0.11 exceeds those of EUA (~ 0.031) and NA (~ 0.027)
12 by a factor of ~ 3 -4. This feature reflects the relatively high rate of emissions over NEC,
13 which results in the highest level of BC deposition over ISCA (Fig. S2a, b). In contrast,
14 being located far from major sources of pollution, the relatively clean Arctic snowpack
15 returns the lowest $\Delta\alpha_{MODIS,corrected}^{LAPs}$ (~ 0.016) of the entire Northern Hemisphere.
16 Consistent with snow albedo reduction, the highest regional-average daily radiative
17 forcing ($RF_{MODIS,daily}^{LAPs}$) occurs in NEC, with values of ~ 12 W m^{-2} , and the lowest
18 regional average occurs in the Arctic, with values of ~ 2.6 W m^{-2} . Regional-average
19 radiative forcing for NA and EUA are both intermediate, with values of ~ 3.1 W m^{-2} and
20 ~ 3.5 W m^{-2} , respectively.

21 On a regional level, NEC $\Delta\alpha_{MODIS,corrected}^{LAPs}$ falls primarily within the range ~ 0.077 –

1 0.14, and intra-regional variability is relatively small due to pervasive heavy pollution
2 (Fig. S2). Compared to snow albedo reduction, the radiative forcing for NEC exhibits
3 a slightly greater spatial variability due to latitude-dependent differences in the flux of
4 surface solar radiances, ranging from $\sim 7.2 \text{ W m}^{-2}$ to $\sim 17 \text{ W m}^{-2}$. In NA, where the
5 principal ISCA are located in southern Canada, the western US, and Central America
6 Plains, $\Delta\alpha_{MODIS,corrected}^{LAPs}$ and $RF_{MODIS,daily}^{LAPs}$ tend to range between $\sim 0.014\text{-}0.046$ and
7 $\sim 1.3\text{-}7.0 \text{ W m}^{-2}$, respectively. In EUA, $\Delta\alpha_{MODIS,corrected}^{LAPs}$ and $RF_{MODIS,daily}^{LAPs}$ fall
8 largely within the respective ranges of $\sim 0.017\text{-}0.049$ and $\sim 1.6\text{-}8.4 \text{ W m}^{-2}$. Central Asia
9 and Mongolia exhibit relatively high values for $\Delta\alpha_{MODIS,corrected}^{LAPs}$ (>0.04) and
10 $RF_{MODIS,daily}^{LAPs}$ ($>2 \text{ W m}^{-2}$), while this pattern likely reflects the influence of
11 anthropogenic BC in addition to natural dust (Pu et al., 2017; Zhou et al., 2019) (Fig.
12 S2a–b).

13 In the Arctic, $\Delta\alpha_{MODIS,corrected}^{LAPs}$ and $RF_{MODIS,daily}^{LAPs}$ both present quite large intra-
14 regional variabilities from ~ 0.0028 to ~ 0.046 and ~ 0.48 to 6.6 W m^{-2} . Greenland has
15 the cleanest snow with $\Delta\alpha_{MODIS,corrected}^{LAPs}$ and $RF_{MODIS,daily}^{LAPs}$ of $\sim 0.011\text{-}0.023$ and
16 $\sim 0.40\text{-}3.3 \text{ W m}^{-2}$. In Canadian Arctic, $\Delta\alpha_{MODIS,corrected}^{LAPs}$ and $RF_{MODIS,daily}^{LAPs}$ are
17 mainly in a range of $\sim 0.012\text{-}0.055$ and $\sim 0.59\text{-}6.1 \text{ W m}^{-2}$. In addition, the relatively high
18 values are found around the edge of ISCA over west of Canadian Arctic. The possible
19 reason is that these areas are suffering from faster snow melting compared with rest of
20 Canadian Arctic in spring, which is characterized by higher snow grain size (Fig. 4b).
21 Hence, more LAPs are accumulated in the surface snow resulting in higher snow albedo

1 reduction. In Russian Arctic, $\Delta\alpha_{MODIS,corrected}^{LAPS}$ and $RF_{MODIS,daily}^{LAPS}$ values increase
2 with altitude present a significant altitude-dependent trend of ~ 0.012 - 0.048 and ~ 1.0 -
3 7.3 W m^{-2} . The snow albedo reduction in eastern Siberia are quite high and comparable
4 with the values in midlatitudes. Moreover, benefiting from the higher solar radiances in
5 eastern Siberia in Spring (Fig. S2d) than that in midlatitudes in Winter-Spring (Fig. 4c
6 and Fig. S2c), $RF_{MODIS,daily}^{LAPS}$ in eastern Siberia is higher than parts of midlatitudes.
7 Even different from the findings in previous modeling studies (e.g. Flanner et al., 2007;
8 2009), the results seem to be comparable with the limited ground-based estimates (Fig.
9 S3). The serious biomass burning in eastern Siberia in Spring may be responsible for
10 such high values (Warneke et al., 2010; Hegg et al., 2009). Overall, the Arctic spatial
11 pattern of $\Delta\alpha_{MODIS,corrected}^{LAPS}$ and $RF_{MODIS,daily}^{LAPS}$ in our study is consistent with the
12 previous studies based on field experiments (Dang et al., 2017) and model simulation
13 (Flanner et al., 2007). Nevertheless, we note that readers should be cautious about our
14 reported high values in Russian Arctic and more field experiments are necessary for
15 validating the results.

16 As mentioned above, the assumption of semi-infinite snowpack will trigger an
17 overestimate for radiative forcing when snow depth is not thick enough. Figure 7 shows
18 the spatial distribution of the ratio of retrieved radiative forcing using semi-infinite
19 snow to radiative forcing using ERA-Interim snow depth. As can be seen that semi-
20 infinite snowpack assumption will lead to an overestimate of up to $\sim 25\%$ in midlatitude
21 areas, where snow depth is thin. In contrast, the influence of snow depth on radiative

1 forcing is negligible in the Arctic, where snow is thick enough to become semi-infinite
2 snowpack. These results demonstrated the important impact of snow depth on radiative
3 forcing retrievals, which must be considered to reduce the overestimate for the
4 following study.

5 **4.5. Accuracy discussion**

6 In spite of the rigorous processes for radiative forcing retrieval, the uncertainty is still
7 existed. For example, light-absorbing particles in the atmosphere will reduce the
8 accuracy of MODIS surface reflectance retrieval, even though the atmospheric
9 correction has been conducted. In addition, previous study pointed out a high scatter
10 when converting NDSI to FSC using Eq. (5), which will induce bias in snow albedo
11 retrieval (Rittger et al., 2013; Riggs et al., 2016). Furthermore, the method for snow
12 grain size retrieval is only based on a single MODIS band at 1.24 μm , which could lead
13 to higher uncertainties. Above all, all of these factors will result in a non-negligible
14 uncertainty for radiative forcing retrieval, which needs to be further discussed.

15 To account for this issue, we consider that the accuracy of atmospheric correction is
16 typically $\pm (0.005 + 0.05 \cdot \text{reflectance})$ under conditions that AOD is less than 5.0 and
17 solar zenith angle is less than 75° according to the MODIS Surface Reflectance User's
18 Guide (Collection 6, <https://modis.gsfc.nasa.gov/data/dataproduct/mod09.php>). In
19 addition, the bias for FSC calculation is assumed as 10% according to Riggs et al.
20 (2016). The bias for snow grain size retrieval is assumed as 30% according to the studies
21 of Pu et al. (2019) and Wang et al. (2017). Figure 8 shows the overall uncertainty of

1 radiative forcing retrieval due to all these factors while Figure S6 show the uncertainty
2 caused by each factor. In general, ~~the the lowerupper (lower) bound and the upper~~
3 ~~bound of the uncertainty positive (negative) uncertainty~~ falls in a range of 15%~108%
4 (-106%~-20%), with atmospheric correction and FSC calculation contributing more to
5 the uncertainty by higher uncertainties than snow grains size retrieval. The highest
6 uncertainty occurs in the Arctic while the lowest uncertainty occurs in NEC.
7 Furthermore, the uncertainty shows a negative correlation with retrieved radiative
8 forcing. The results indirectly demonstrated the reasonability of different correction
9 factors performed in different regions. For example, the value of 1.6 used in NEC
10 suggests that the correction approach works well for heavily polluted snow, while the
11 value of 6.0 used in Greenland for relatively clean snow suggests that the method
12 becomes not accurate enough.

13 It worth noting that the uncertainties from these factors could not fully explain the high
14 correction factor in clean snow. The reason for why the ratio $\Delta\alpha_{MODIS,daily}^{LAPs}/$
15 $\Delta\alpha_{in-situ,daily}^{LAPs}$ to be larger than 1 is mostly like that There are four probable reasons: (1)
16 the effect of snow surface roughness (Manninen et al., 2020) and vegetation (Pu et al.,
17 2019), which were without regarding in SNICAR, definitely probably reduce the
18 derived albedo from MODIS and therefore result in overestimate of the albedo
19 reduction attributed to LAPs. Moreover, there are other potential factors causing errors:
20 the rough snow surface and uncertainties of vegetation and soil reflectance can
21 effectively influence radiative foreing retrieval; (21) MODIS has variably spaced and

1 discrete spectral bands and thus cannot provide a continuous spectral measurement of
2 reflectance. This results in a non-negligible uncertainty in retrieving the radiative
3 forcing by LAPs in snow.~~MODIS cannot proceed with a continuous spectral~~
4 ~~measurement of a continuous variable forcing like what LAPs afford to snow albedo~~
5 ~~due to the variably spaced and discrete bands of MODIS, which prevents a more~~
6 ~~quantitative retrieval and thus results in a non-negligible uncertainty in radiative forcing~~
7 ~~retrieval~~ (Painter et al., 2012); (32) We use the retrieved radiative forcing in a pixel size
8 of $0.05^\circ \times 0.05^\circ$ to compare with the in-situ radiative forcing calculated from the
9 measured BC_{equiv} concentration with a sample site located somewhere within the
10 pixel~~the sample site located in the center of the pixel~~. However, such a comparison may
11 not be representative ~~true~~ at some sites due to the inhomogeneous spatial distribution
12 of LAP contents, which will influence radiative forcing retrieval; (43) In-situ
13 measurements also have uncertainties, which may cause a high bias for snow albedo
14 reduction in clean snow. For example, a 10% bias for 50 ng g^{-1} BC can result in an 8%
15 bias for snow albedo reduction.

16 **4.6. Attribution to the spatial variability of snow albedo reduction and radiative** 17 **forcing**

18 Here, we address the attributions to the spatial variability of snow albedo reduction and
19 radiative forcing. As discussed in Sect. 3.2.5, the spatial variability in snow albedo
20 reduction and radiative forcing are largely dependent on LAP content, snow grain radius,
21 snow depth, and the geographic factor. Figure 9 illustrates the fractional contributions

1 of each factor within the study regions. For the Northern Hemisphere ISCA as a whole,
2 LAPs (I_{LAPs}) is the greatest contributor (84.3%) to snow albedo reduction, followed by
3 SD (13.7%); R_{eff} and G have only a minor influence (1.9% and <1%, respectively)
4 (Fig. 9a). This result confirms that the concentration of LAPs in the snowpack plays a
5 fundamental role in spatial variability of snow albedo reduction.

6 LAPs also constitute the dominant contributors to snow albedo reduction on a regional
7 scale, accounting for 96.0% of the Arctic signal and 56.7% in EUA and 49.9% in NA,
8 and are the second largest contributor in NEC (40.3%). The contribution of SD is
9 greatest in NEC (56.3%), with slightly lower values in EUA (40.3%) and NA (48.8%),
10 reflecting the significant spatial variability in SD across these regions. In the Arctic,
11 the snowpack is sufficiently thick to be considered a homogeneous, semi-infinite
12 snowpack and thus the contribution of SD is negligible. In contrast, R_{eff} makes only
13 minor contributions in NEC (3.3%), NA (1.3%), EUA (2.8%) and the Arctic (1.4%).
14 Finally, G makes the smallest contribution to snow albedo reduction (<1%), both on
15 regional and global scales.

16 On a hemispheric scale, the greatest contributors to radiative forcing are LAP content
17 (70.0%) and G (22.3%), followed by SD (7.6%). As with snow albedo reduction,
18 R_{eff} plays only a minor role. The influence of G on spatial variability in radiative
19 forcing is attributed to the high degree of variability in latitude-dependent solar
20 ~~radiance~~ radiative fluxes among ISCA. On a regional scale, the respective
21 contributions of LAP content, G , and SD are also comparable among the four study

1 areas, accounting for 34.1%, 11.1%, and 52.0% of radiative forcing in NEC, 39.2%,
2 13.9%, and 46.4% in NA, and 48.0%, 19.3%, and 31.6% in EUA. The Arctic radiative
3 forcing is dominated by LAPs (85.6%) and G (12.7%).

4 In summary, LAPs play a dominant role in the spatial variability of snow albedo
5 reduction and radiative forcing. Our results also highlight the significant contribution
6 of SD to snow albedo reduction and G to radiative forcing.

7 **4.7. Comparisons with model simulations**

8 To investigate the global distribution and variance of RFLS, previous studies have
9 tended to rely on Earth system models with minimal cross-checking from in-situ
10 measurements or remote sensing observations (Qian et al., 2015; Skiles et al., 2018). In
11 this study, we compared MODIS retrievals with CESM2 to improve our understanding
12 of the magnitude of RFLS on a global scale.

13 Employing snow BC concentrations from CESM2, we also calculated December-May
14 daily radiative forcing (RF_{CESM2}) for the Northern Hemisphere ISCA during the period
15 2003–2014 (Fig. 10a). Statistics are presented in Fig. S7. Briefly, RF_{CESM2} exhibits
16 strong spatial inhomogeneity, with values ranging from 0.20 W m^{-2} to 5.6 W m^{-2} . The
17 highest regional average in RF_{CESM2} occurs in NEC ($\geq 10 \text{ W m}^{-2}$) and the lowest in
18 the Arctic ($\leq 0.5 \text{ W m}^{-2}$), consistent with $RF_{MODIS,daily}^{LAPs}$.

19 Figure 10b depicts the comparison of $RF_{MODIS,daily}^{LAPs}$ and RF_{CESM2} . In NEC, RF_{CESM2}
20 (15 W m^{-2}) compares well with $RF_{MODIS,daily}^{LAPs}$ (12 W m^{-2}), with a significant

1 correlation at the 99% confidence level. For EUA, RF_{CESM2} (3.8 W m^{-2}) is similar to
2 $RF_{MODIS,daily}^{LAPs}$ (3.5 W m^{-2}). For NA, RF_{CESM2} (1.2 W m^{-2}) is lower than
3 $RF_{MODIS,daily}^{LAPs}$ (3.1 W m^{-2}) and the spatial correlation between them are poor. In the
4 Arctic, RF_{CESM2} is correlated with $RF_{MODIS,daily}^{LAPs}$ at the 99% confidence level.
5 However, RF_{CESM2} (1.7 W m^{-2}) is lower than $RF_{MODIS,daily}^{LAPs}$ (2.6 W m^{-2}) by a factor
6 of 1.5.

7 Overall, the RFLS derived from our MODIS retrievals and modeling-based estimates
8 exhibit a same magnitude over the Northern Hemisphere. In NEC, the MODIS- derived
9 and model-derived estimates show good general agreement, indicating the satisfactory
10 performance of ~~CESM2~~ Earth system modeling in this heavily polluted region. In EUA,
11 average radiative forcing values are comparable but the spatial correlation is relatively
12 poor, while MODIS retrievals for the Arctic are significantly higher than those
13 simulations.

14 **5. Discussion**

15 In recent decades, there has been increasing scientific interest in snow LAPs due to
16 their role in the climate system, and numerous studies have attempted to evaluate RFLS.
17 In addition to making global-scale comparisons between our MODIS retrievals and
18 model-based estimates, this study collects a comprehensive set of radiative forcing
19 estimates, based on local-scale observations and remote sensing, to make quantitative
20 regional- and global-scale comparisons and synthetically evaluate the magnitude of
21 RFLS (Table 2). This approach also affords the opportunity to examine the MODIS

1 retrievals used in our study.

2 Dang et al. (2017) reported RFLS values of 7–18 W m⁻², 0.6–1.9 W m⁻², and 0.1–0.8
3 W m⁻² for northern China, North America, and the Arctic, respectively, which only
4 focused on the period of January-March, and therefore are smaller than our retrievals.
5 In NA, Sterle et al. (2013) estimated a daily-averaged RFLS of ~2.5-40 W m⁻² for the
6 eastern Sierra Nevada in February-May, 2009, while Miller et al. (2016) reported a daily
7 RFLS of ~35-86 (37-100) W m⁻² based on in-situ measurements (remote sensing) in
8 the San Juan Mountains in May 2010. Both values are higher than our estimate (~3.1
9 W m⁻²), potentially due to the significant dust deposition in those areas.

10 We also collected the average-daily RFLS simulated by regional and/or global climate
11 models (Table 2). For NEC, Zhao et al. (2014) and Qian et al. (2014) reported values
12 of 10 W m⁻² in January-February and 5–10 W m⁻² in April, respectively. In NA, Qian
13 et al. (2009) provided an estimate of 3–7 W m⁻² for the central Rockies and southern
14 Alberta in March, while Oaida et al. (2015) reported an average RFLS of 16 W m⁻²
15 over the western US in spring. Finally, Qian et al. (2014) and Qi et al. (2017) estimated
16 RFLS values of <0.3 W m⁻² and 0.024–0.39 W m⁻² for the Arctic in April, respectively.

17 We consider our retrievals for NEC to be comparable with these regional model
18 simulations, despite some disparity. However, we note that our result is significantly
19 lower than those of previous studies in NA, but higher in the Arctic.

20 On a global scale, Hansen and Nazarenko (2004) reported the RFLS is 0.3 W m⁻², while
21 Flanner et al. (2007) showed a RFLS of ~0.05 W m⁻². For the North Hemisphere as a

1 whole, Bond et al. (2013) estimated a climate forcing of 0.13 W m^{-2} . Each of these
2 previous values is significantly lower than our retrieval ($\sim 2.9 \text{ W m}^{-2}$). However, those
3 studies included all areas regardless of snow covered throughout the whole year, while
4 our results are only for Northern Hemisphere ISCA from December to May.

5 Overall, we consider our MODIS-based retrievals to be physical realistic on both
6 regional and global scales, although we note a number of differences between our
7 results and those generated by different methods. On the other hand, while in-situ
8 measurements are the most precise, their spatial coverage is restricted by logistical
9 limitations and the extreme environments involved. Conversely, models can provide
10 broad perspectives of climatic impacts yet are typically undermined by large uncertainty.
11 Therefore, we argue that remote sensing provides a powerful technique, with high
12 spatial and temporal resolutions, that can bridge the gap between in-situ measurements
13 and climate models and reduce the uncertainties associated with the latter. Further
14 retrieval of remote-sensing data, including the use of multiple satellites and sensors, is
15 therefore warranted to exploit this opportunity fully. We also indicate the fact that parts
16 of central EUA and Russian Arctic, however, studies are barely performed but desired.
17 Finally, we note that in-situ observations remain limited, and more field campaigns are
18 needed to constrain remote sensing retrievals and modeling simulations.

19 **6. Conclusion**

20 We presented a global-scale evaluation of the daily radiative forcing of LAPs in the
21 Northern Hemisphere snowpack (RFLS), estimated from remote-sensing data. The

1 satellite-retrieved RFLS also has implications for expanding the value of limited in-situ
2 measurements, which can provide valuable information for climate models and help
3 optimize model simulations.

4 Based on the corrected snow albedo reduction ($\Delta\alpha_{MODIS,corrected}^{LAPs}$), we calculated
5 average-daily RFLS ($RF_{MODIS,daily}^{LAPs}$) during December-May for the period 2003–2018.

6 For the identified snow covered area over Northern Hemisphere as a whole, average
7 $\Delta\alpha_{MODIS,corrected}^{LAPs}$ is ~ 0.021 and $RF_{MODIS,daily}^{LAPs}$ is $\sim 2.9 \text{ W m}^{-2}$. We also observed
8 distinct spatial variability in snow albedo reduction and RFLS. The highest regional-
9 average $\Delta\alpha_{MODIS,corrected}^{LAPs}$ (~ 0.11) and $RF_{MODIS,daily}^{LAPs}$ ($\sim 12 \text{ W m}^{-2}$) occur in
10 northeastern China, while the lowest regional averages of ~ 0.016 and $\sim 2.6 \text{ W m}^{-2}$,
11 respectively, are observed in the Arctic. Moreover, we indicated that the semi-infinite
12 assumption could overestimates up to $\sim 25\%$ of RFLS, especially for thin and patchy
13 snow, such as midlatitudes in Eurasia and NA. In addition, if the ground-based
14 corrections were not considered, the total uncertainty of RFLS retrievals is in the range
15 of $15\% \sim 108\%$ ($-106\% \sim -20\%$) due to atmospheric correction, snow cover fraction
16 calculation and snow grain size retrieval.

17 Following this assessment, we made quantitative attributions of the spatial variability
18 in snow albedo reduction and radiative forcing. Our results indicate that the LAP
19 content is the largest contributor (84.3%) to spatial variance in snow albedo reduction,
20 followed by snow depth (13.7%), whereas snow grain size (1.9%) and the geographic
21 factor G ($<1\%$) are only minor contributors on a Northern Hemispheric scale. LAP

1 content and G account for 70.0% and 22.3% of the spatial variability of radiative
2 forcing, respectively, following by SD (7.6%) over Northern Hemisphere.

3 Retrieved RFLS values are compared spatially with the model-derived estimates of the
4 CESM2. Our results indicate that MODIS retrievals show the same magnitude with
5 modeled estimates for Northern Hemisphere. However, although the [CESM2Earth](#)
6 [system models](#) perform well in NEC, there remain large uncertainties in the Arctic. To
7 evaluate and examine the MODIS retrievals synthetically, we then compared the
8 retrieved RFLS to previously published estimates, including local-scale observations,
9 remote sensing retrievals, and regional- and global-scale model simulations. The results
10 of this evaluation suggest that MODIS retrievals are generally realistic, despite a
11 number of important differences among the various methods.

12 Finally, we urge the community to expand the ground-based measurements of the global
13 snowpack, particularly in those regions currently lacking in-situ observations. Such
14 development would help further constrain and improve satellite-based retrievals in the
15 future. We propose that climate models validated by these refined remote sensing
16 retrievals should be able to capture the RFLS more accurately, thereby providing more
17 reliable estimates of the future impacts of global climate change.

1 **Data availability.**

2 MODIS data can be found at <https://earthdata.nasa.gov/> (last access: 20 January 2019).

3 CERES data can be found from NASA's Clouds and the Earth's Radiant Energy System

4 at <https://ceres.larc.nasa.gov> (last access: 12 April 2019). Shuttle Radar Topography

5 Mission (SRTM) digital elevation data are provided by the US Geological Survey at

6 <https://www.usgs.gov/> (last access: 9 December 2018). Snow depth can be found from

7 ERA-Interim at <https://www.ecmwf.int> (last access: 15 January 2019). BC emission

8 data can be found at <http://inventory.pku.edu.cn> (last access: 5 June 2019). BC

9 deposition data can be found at <https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/> (last

10 access: 5 June 2019). CMIP6 data can be found at <https://esgf-node.llnl.gov/> (last access:

11 15 July 2019). Surface measurement datasets are from Wang et al. (2013, 2017), Ye et

12 al. (2012) and Doherty et al. (2010, 2014). Springtime radiative forcing due to LAPs in

13 snow is derived from a GCM run by Flanner et al. (2007).

1 **Author contributions.**

2 PW and WX designed the study and evolved the overarching research goals and aims.
3 CJC carried the study out and wrote the first draft with contributions from all co-authors.
4 CJC and STL applied formal techniques such as statistical, mathematical and
5 computational to analyze study data. ZY prepared input data and managed activities to
6 annotate, scrub data and maintain research data. WDY completed the implementation
7 of the computer code and supporting algorithms used for the calculations in this study.
8 PW and WX assumed oversight and leadership responsibility for the research activity
9 planning and execution. All authors contributed to the improvement of results and
10 revised the final paper.

1 **Competing interests.**

2 The authors declare that they have no conflict of interest.

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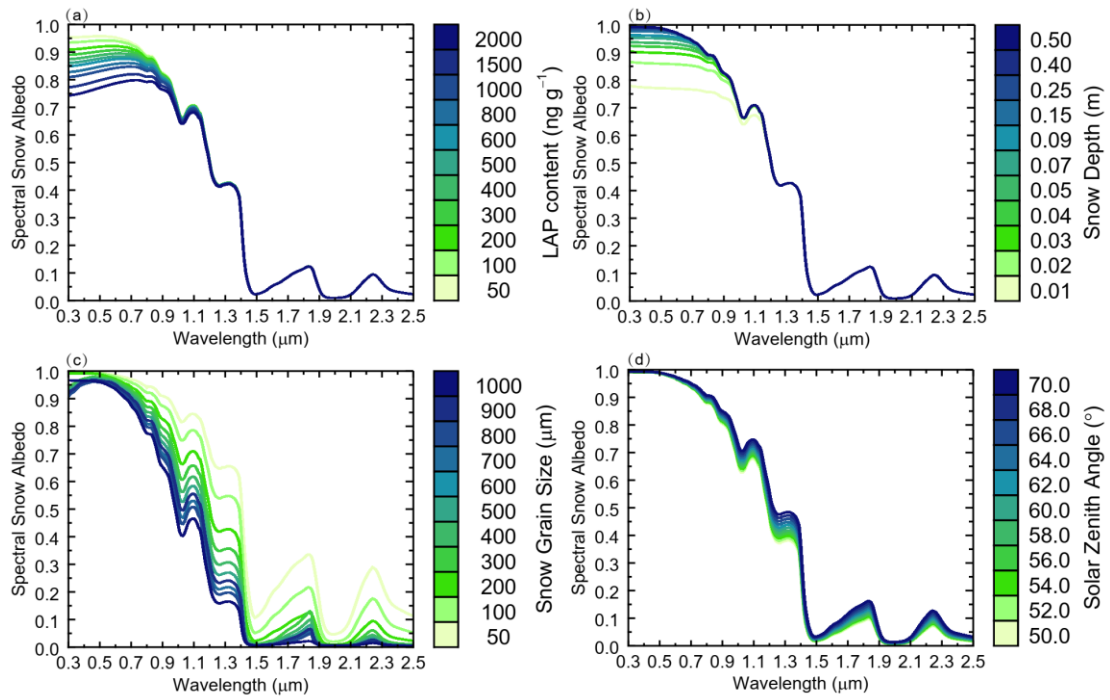
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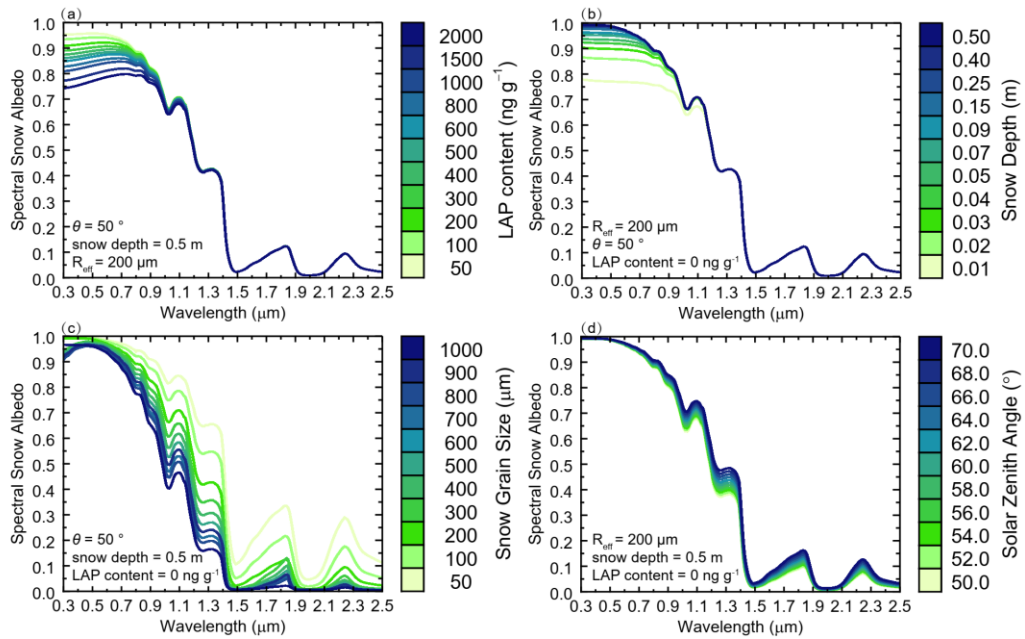
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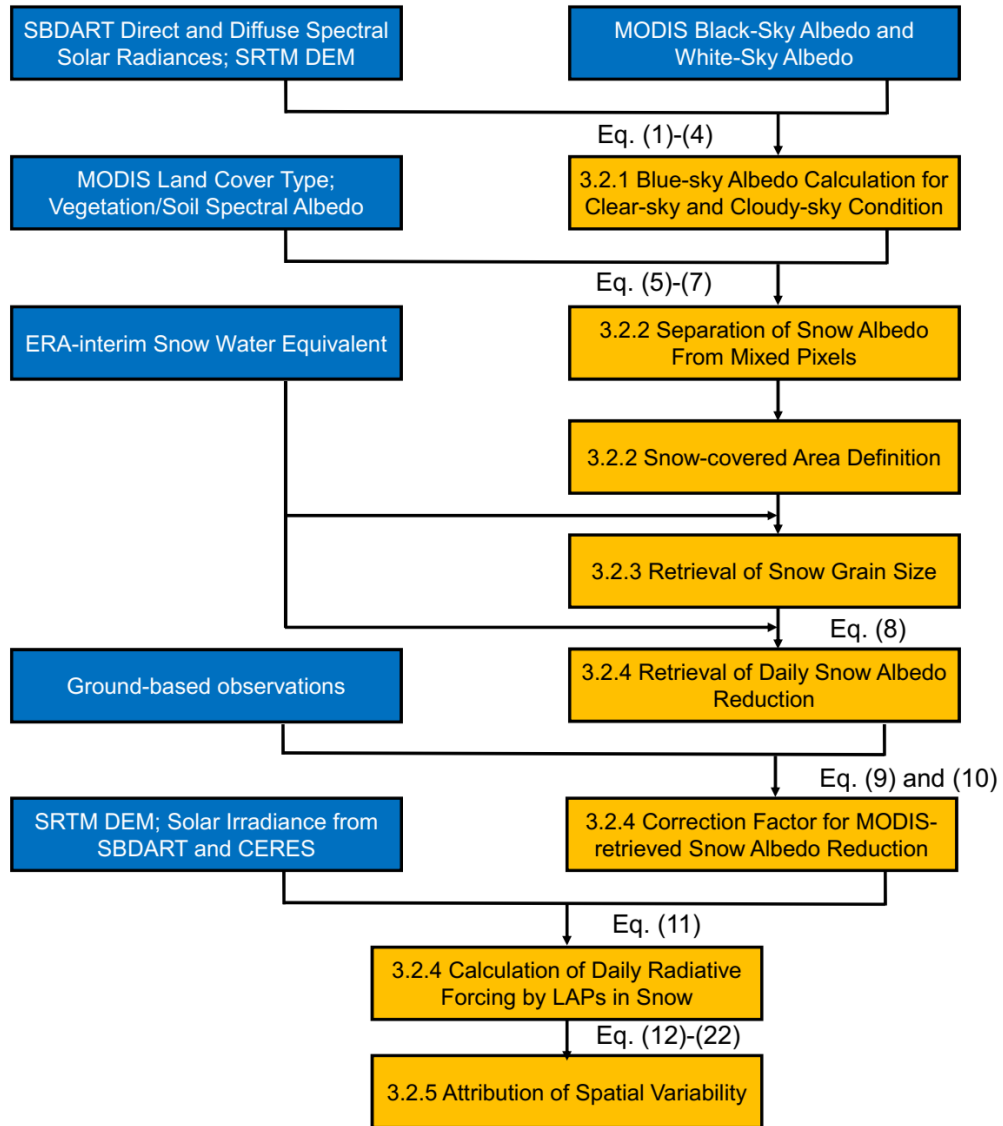


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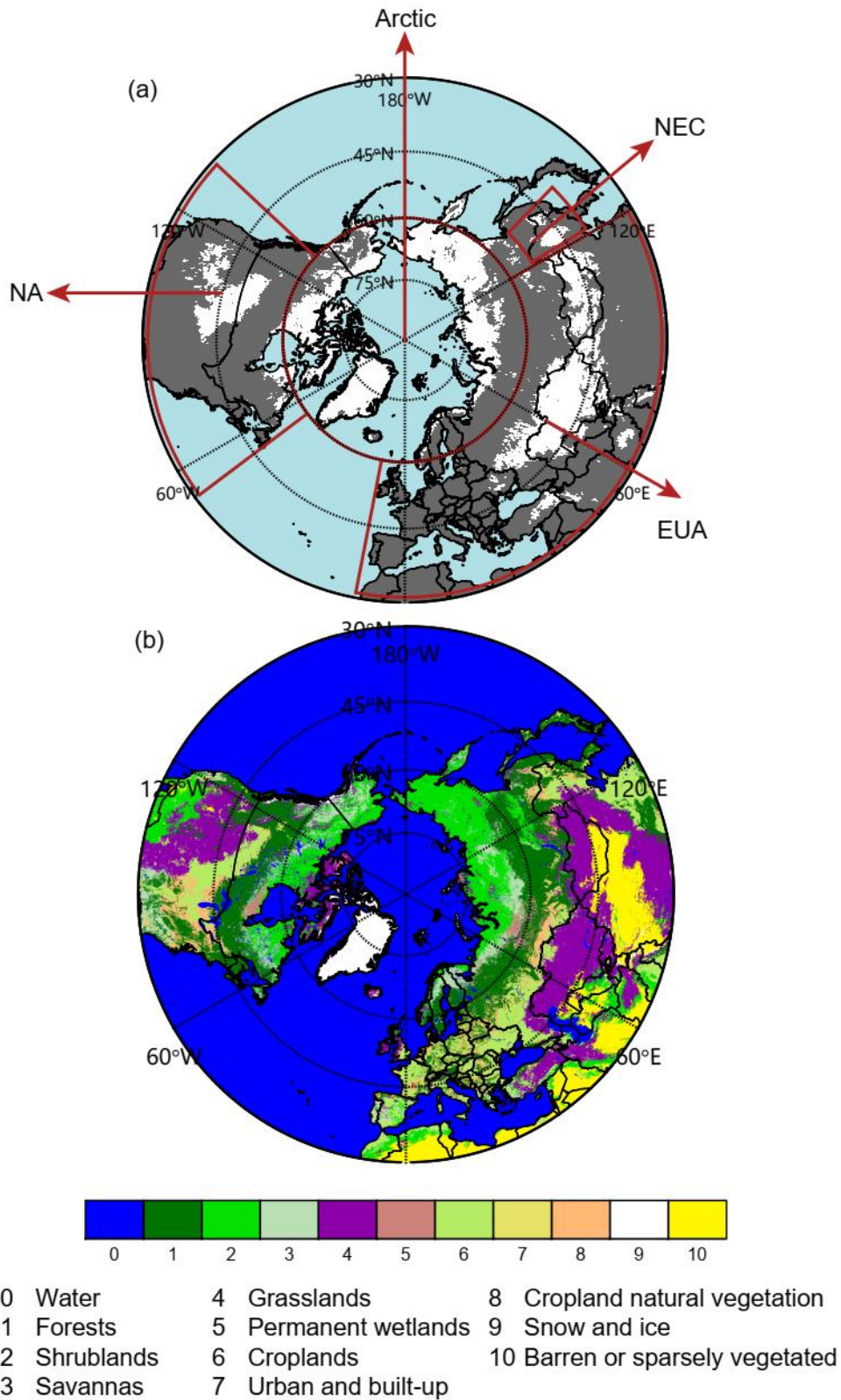
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3 Figure 1. Variations in spectral snow albedo due to (a) LAP content (ng g^{-1}), (b) snow depth (m), (c)
 4 snow grain size (μm), and (d) solar zenith angle (deg.) while other three parameters are kept
 5 constant.



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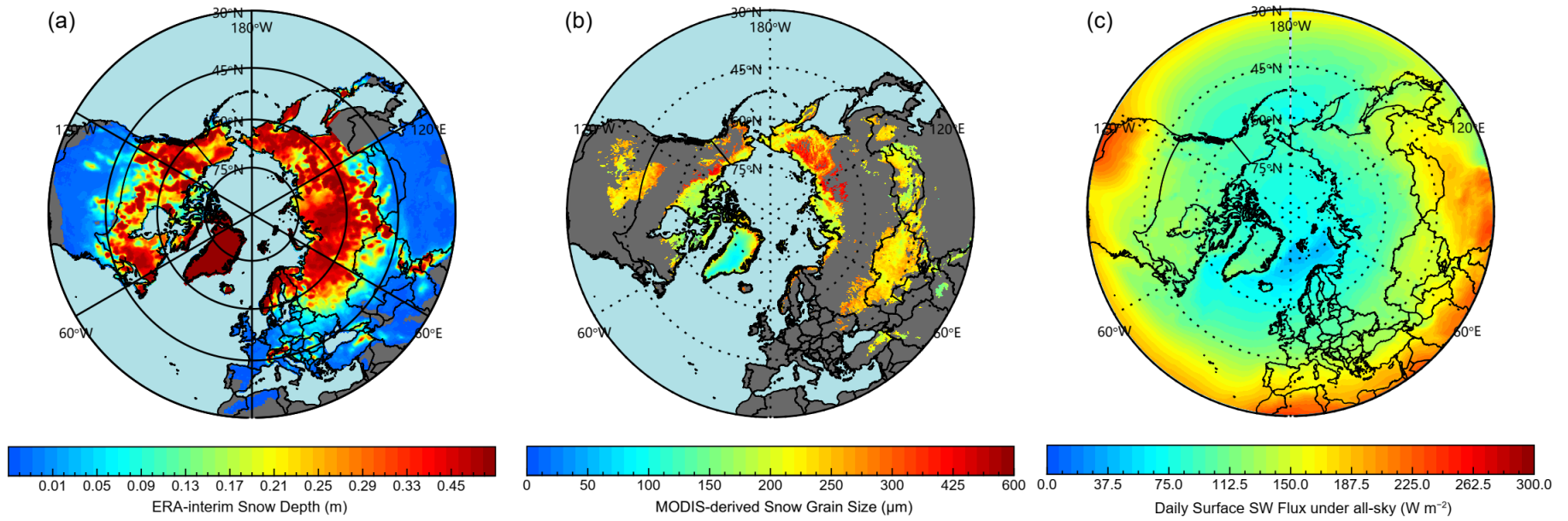
2 Figure 2. Workflow depicting the calculation and validation of radiative forcing of Laps in snow:
 3 the blue boxes denote the external input data, while the orange boxes are used for calculations in
 4 this study.



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2 Figure 3. Spatial distributions of (a) identified snow-covered areas (ISCA) and (b) the different land-
 3 cover types, based on MODIS data, for the Northern Hemisphere. ISCA (white) can be separated
 4 into northeastern China (NEC), Eurasia (EUA), North America (NA), and the Arctic.

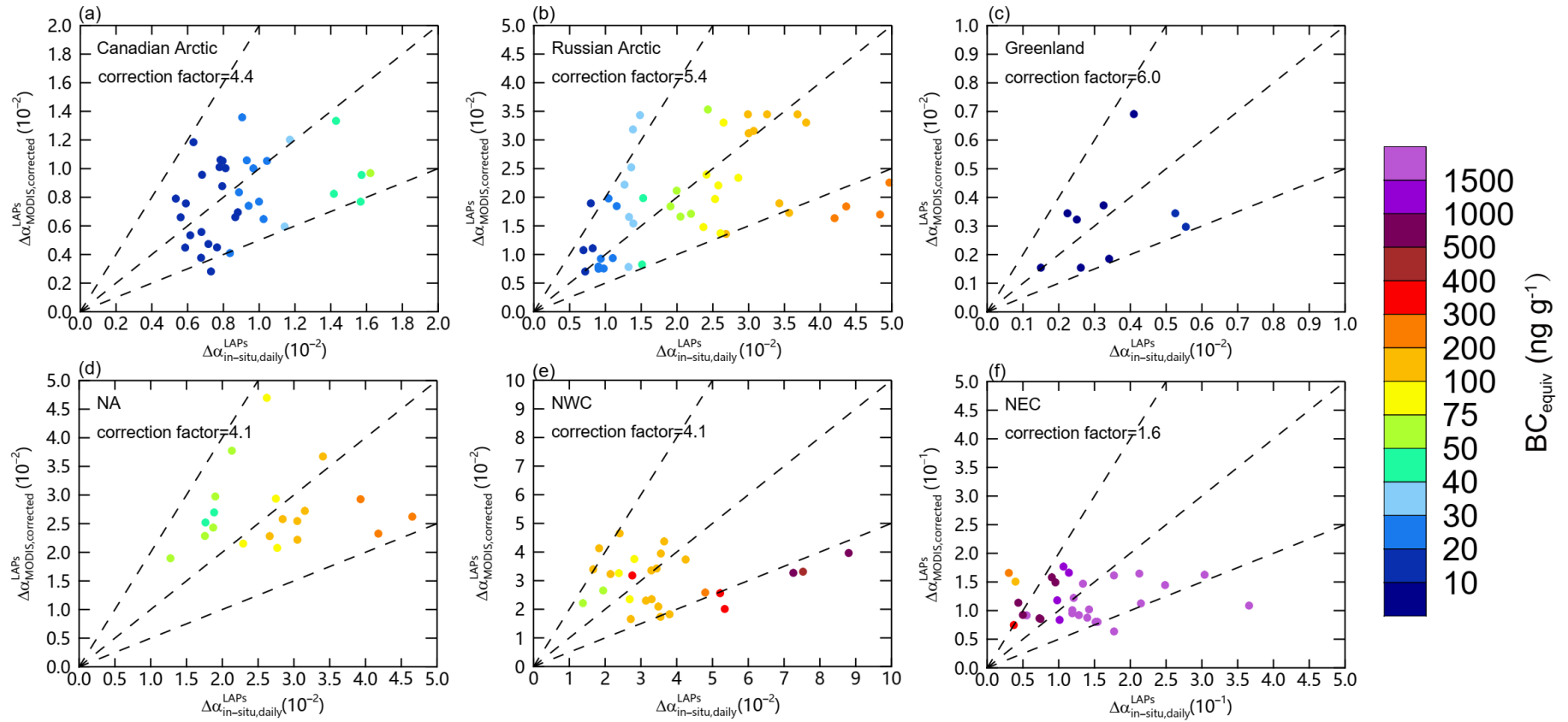
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3 Figure 4. Spatial distributions of 2003-2018 averaged (a) snow depth from ERA-interim, (d) snow grain size retrieved by MODIS, and (c) total downward shortwave
4 flux at the surface during December-May from CERES.

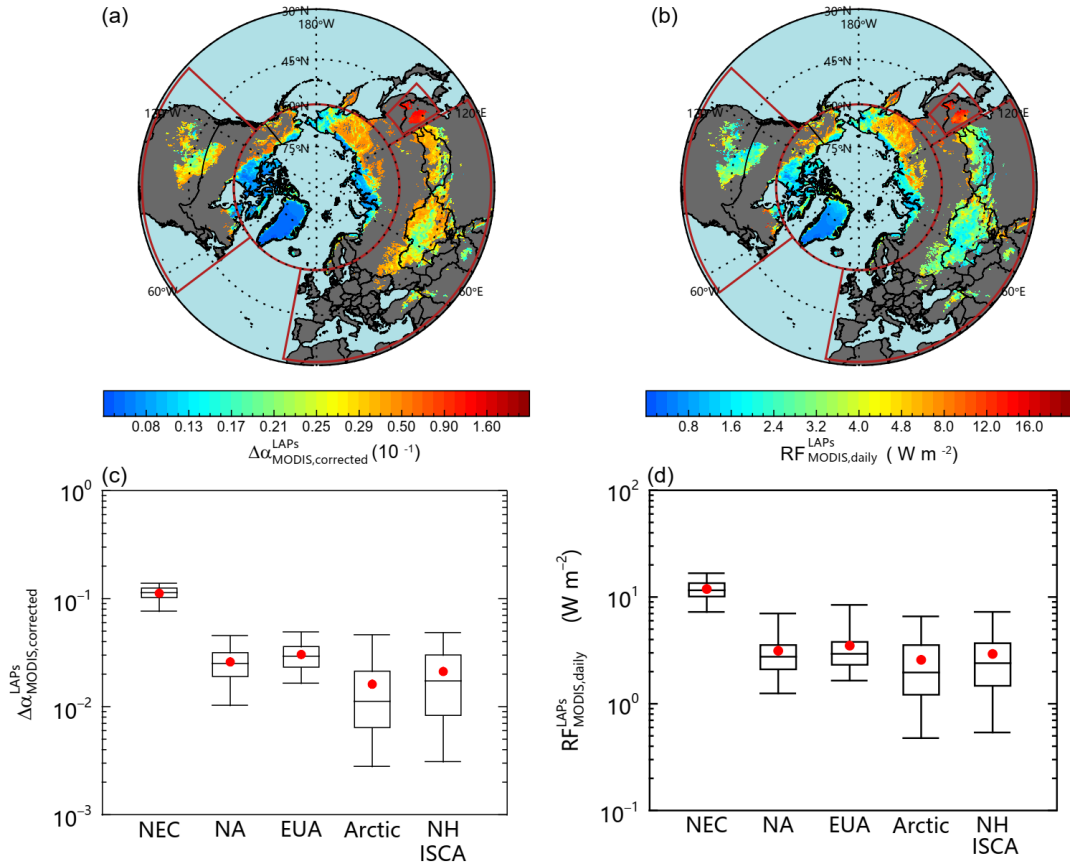
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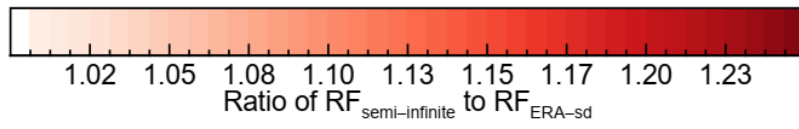
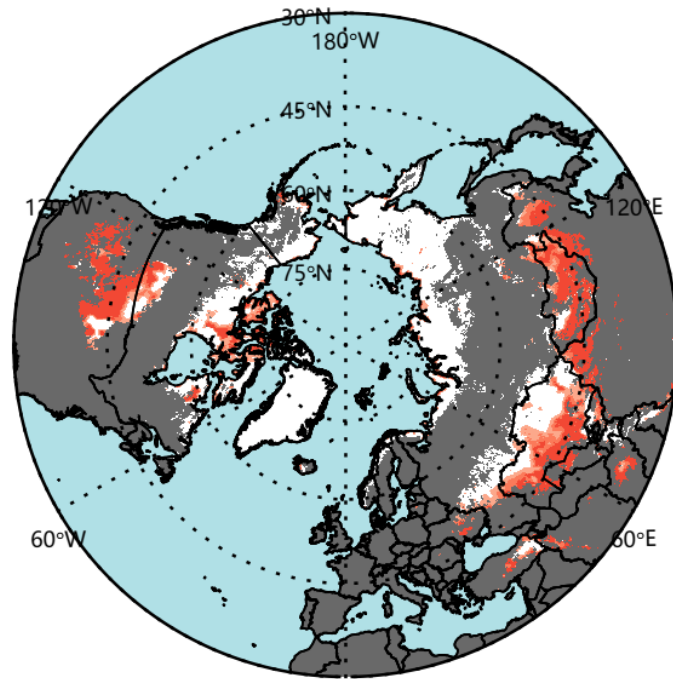
3 Figure 5. Scatterplots of $\Delta\alpha_{MODIS,corrected}^{LAPS}$ versus $\Delta\alpha_{in-situ,daily}^{LAPS}$. Panels (a)–(f) represent the snow samples collected in Canadian Arctic, Russian Arctic, Greenland,
 4 North America, Northwestern China, and Northeastern China, respectively.

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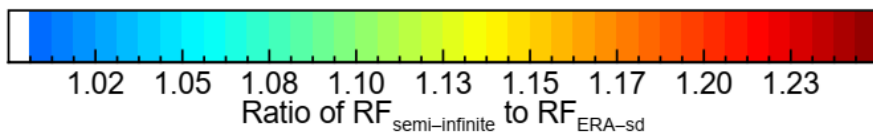
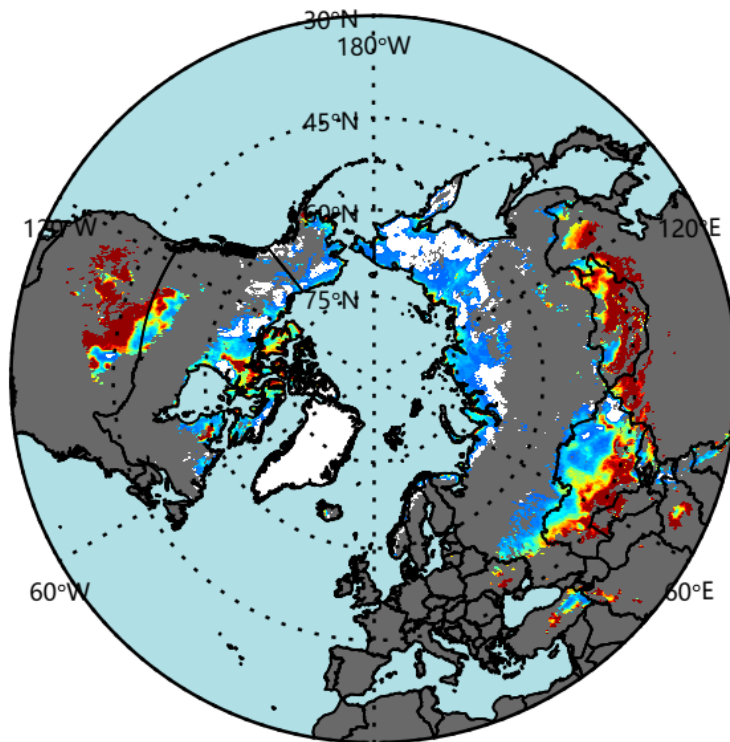


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3 Figure 6. Spatial distributions of averaged (a) $\Delta\alpha_{MODIS,corrected}^{LAPs}$, (b) $RF_{MODIS,daily}^{LAPs}$ and statistics
 4 for regionally averaged (c) $\Delta\alpha_{MODIS,corrected}^{LAPs}$ and (d) $RF_{MODIS,daily}^{LAPs}$ for the Northern Hemisphere
 5 ISCA in December-May during the period 2003–2018. The boxes denote the 25th and 75th quantiles,
 6 and the horizontal lines represent the 50th quantiles (medians), the averages are shown as red dots;
 7 the whiskers denote the 5th and 95th quantiles.

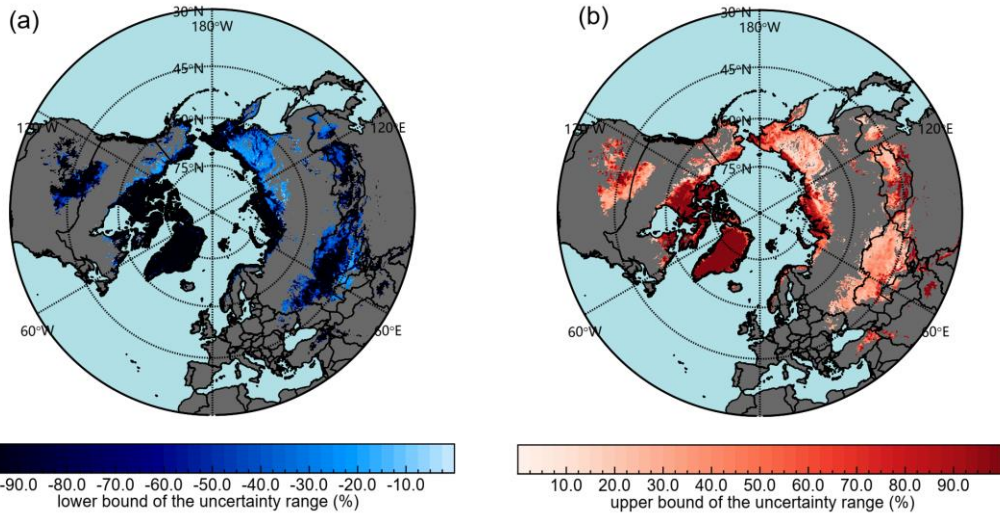
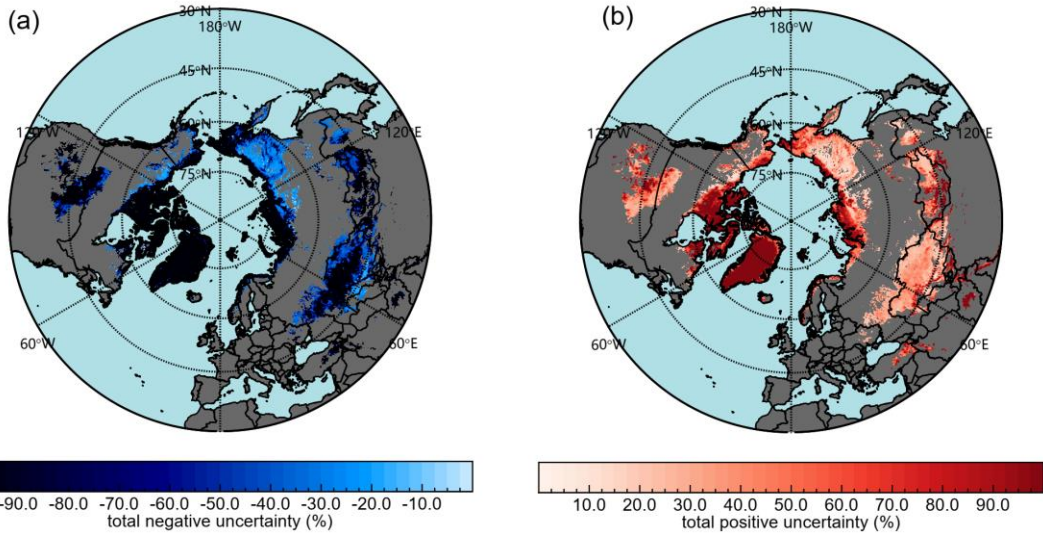


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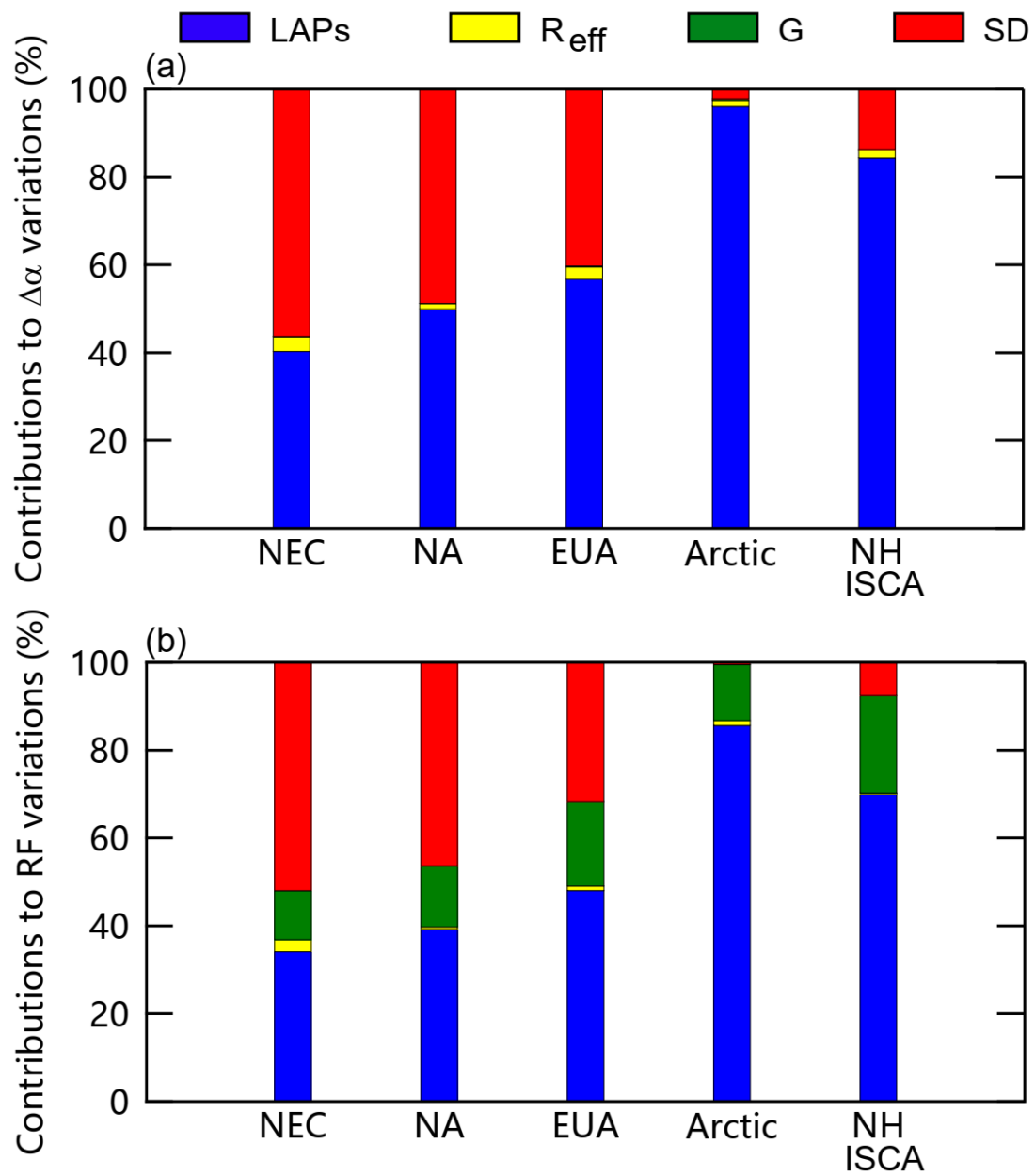


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2 Figure 7. The spatial distribution of the ratio of retrieved radiative forcing using semi-infinite snow
 3 to radiative forcing using ERA-Interim snow depth.

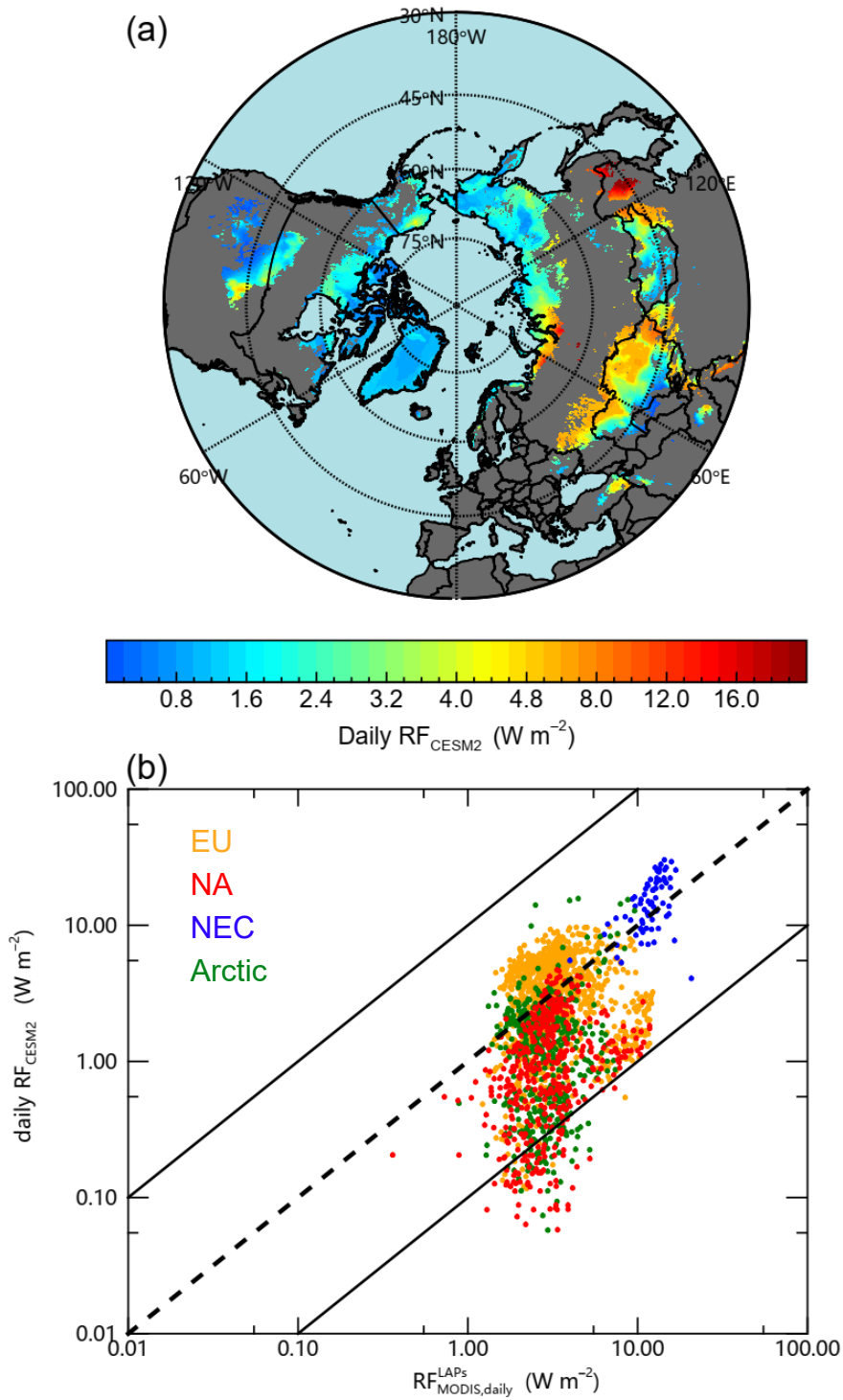


3 Figure 8. The overall lower bound and upper bound of the uncertainty range ~~uncertainty~~ of radiative
 4 forcing retrieval due to atmospheric correction, MODIS-derived snow grain size retrieval and snow
 5 cover fraction calculation.



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2 Figure 9. Fractional contributions of LAPs, snow grain size (R_{eff}), geographic factor (G), and snow
 3 depth (SD) to the spatial variations of (a) snow albedo reduction and (b) daily radiative forcing.



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2 Figure 10. (a) Spatial distributions of average-daily radiative forcing (RF_{CESM2}), based on the
 3 CESM2 soot content of snow in December-May for the period 2003–2014. (b) Scatterplot of

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$RF_{MODIS,daily}^{LAPs}$ versus RF_{CESM2} .

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Table 1. Statistics for regionally averaged (5th and 95th quantiles) albedo reduction ($\Delta\alpha_{MODIS,corrected}^{LAPs}$) and daily radiative forcing ($RF_{MODIS,daily}^{LAPs}$, $W m^{-2}$)

	Northeastern China	EUA	NA	Canadian Arctic	Greenland	Russian Arctic	ISCA over Northern Hemisphere
Albedo reduction ($\Delta\alpha_{MODIS,corrected}^{LAPs}$)	0.11 (0.077~0.14)	0.031 (0.017~0.049)	0.027 (0.014~0.046)	0.025 (0.012~0.055)	0.016 (0.011~0.023)	0.028 (0.012~0.048)	0.021 (0.0031~0.049)
Daily radiative forcing ($RF_{MODIS,daily}^{LAPs}$, $W m^{-2}$)	12 (7.2~17)	3.5 (1.6~8.4)	3.1 (1.3~7.0)	2.6 (0.59~6.1)	1.3 (0.40~3.3)	3.3 (1.0~7.3)	2.9 (0.54~7.3)

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Table.2 Comparisons of radiative forcing due to LAPs in snow (this study) with observed and model-simulated values from previous studies

Study	Region	Time period	Method	Radiative forcing (W m^{-2})
Miller et al. (2016)	San Juan Mountains	May, 2010	Remote sensing	~37-100
Sterle et al. (2013)	eastern Sierra Nevada	Feb to May, 2009	In-situ measurements	~2.5-40
Miller et al. (2016)	San Juan Mountains	May, 2010	In-situ measurements	35-86
Dang et al. (2017)	Northern China	Jan and Feb, 2010 and 2012	In-situ measurements	7-18
	North America	Jan-Mar, 2013-2014	In-situ measurements	0.6-1.9
	The Arctic	Spring, 2005-2009	In-situ measurements	0.1-0.8
Hansen and Nazarenko (2004)	North Hemisphere		Model simulations	0.3
Qian et al. (2009)	western United States	Mar	Model simulations	~3-7
Bond et al. (2013)	Global	industrial era	Model simulations	0.13
Flanner et al. (2007)	Global	Annual 1998 (strong)	Model simulations	0.054
		Annual 2001(weak)		0.049
Qian et al. (2014)	Northeastern China	Apr	Model simulations	5-10
	North America	Apr	Model simulations	2-7

	The Arctic	Apr	Model simulations	<0.3
Zhao et al. (2014)	Northeastern China	Jan and Feb, 2010	Model simulations	10
Oaida et al. (2015)	western US	Spring, 2009-2013	Model simulations	16
Qi et al. (2017)	The Arctic	Apr, 2008	Model simulations	0.024-0.39
This study	Northeastern China	Dec-May, 2003-2018	Remote sensing	12
	NA			3.1
	Canadian Arctic			2.6
	Russian Arctic			3.3
	Greenland			1.3
	EUA			3.5
