Oct10, 2017

Dear editor,

The manuscript: *Black carbon and mineral dust in snow cover on the Tibetan Plateau* by Zhang Y.L. et al.

The manuscript number: tc-2017-111

We greatly appreciate the reviewers' constructive comments to improve the paper. We have revised our manuscript according to these comments (blue color in the main text), and hope the revised manuscript is suitable for publication in The Cryosphere.

The "point to point" response to comments are listed as below.

Sincerely yours, Yulan Zhang and Shichang Kang

Response to Referee #1

General opinion: The authors report the observation of light-absorbing impurities in snow over Tibetan Plateau. Based on these field observed data, they calculated the albedo reduction induced by black carbon and mineral dust, and the corresponding impact on snow energy budget. The field data reported in this work are valuable for quantifying the impact of light-absorbing particles on snow albedo and the analysis based on this field data are informative. However, some discussion and conclusions given in this paper are not accurate and need to be modified. This manuscript also contains a lot of types/grammar errors (some obvious errors are listed below) that need to be corrected before this manuscript can be considered for publication. Here are the suggestions/comments that the authors may find useful.

Answer: Thank you very much for the comments. We have carefully revised according to the comments. The manuscript has also been improved by a native speaker to reduce the types/grammar errors.

Comments and Questions

Page 1 Line 30-32: BC is recognized as an important climate forcer not only because it absorbs sunlight, but also because a large fraction of BC are emitted from anthropogenic sources. Please be more accurate at here.

Answer: We have revised as following in the main text (Page 1-2):

Atmospheric BC is a distinct type of carbonaceous material from incomplete combustion of biomass/biofuel and fossil fuel. A large fraction of BC is emitted from anthropogenic activities

(Bond et al., 2013). Because BC can absorb solar radiation, influence cloud processes, and alter the melting of snow cover and glaciers, it has been considered to be the second most important climate forcer in the Earth's climate system only after carbon dioxide (Andreae and Ramanathan, 2013; Bond et al., 2013; Ramanathan and Carmichael, 2008).

References:

Andreae, M. O., and Ramanathan, V.: Climate's dark forcings, Science, 340, 280–281, 2013.

- Bond, T. C., Doherty, S. J., Fahey, D. W., Forster, P. M., Berntsen, T., DeAngelo, B. J., Flanner, M. G., Ghan, S., Kärcher, B., Koch, D., Kinne, S., Kondo, Y., Quinn, P. K., Sarofim, M. C., Schultz, M. G., Schultz, M., Venkataraman, C., Zhang, H., Zhang, S., Bellouin, N., Guttikunda, S.K., Hopke, P. K., Jacobson, M. Z., Kaiser, J. W., Klimont, Z., Lohmann, U., Schwarz, J. P., Shindell, D., Storelvmo, T., Warren, S. G., Zender, C. S.: Bounding the role of black carbon in the climate system: A scientific assessment, J. Geophys. Res. Atmos., 118, 5380–5552, doi:10.1002/jgrd.50171, 2013.
- Ramanathan, V. and Carmichael, G.: Global and regional climate changes due to black carbon, Nat. Geosci., 1, 221-227, doi:10.1038/nego156, 2008.

Page 2 Line 16: ": : : snow covered range" ! snow covered region? Answer: It is "snow covered region". (Page 2, Line 26)

Line 20: "Confirming radiative transfer modeling". It is not very clear to readers what does the authors mean. Please consider revise this.

Answer: The sentence has been revised as following (Page 2, Line 23-31):

In the European Alps, observed reflectance provided evidence that seasonal input of dust can strongly decrease the spectral properties of snow, in particular, from 350 to 600 nm (Di Mauro et al., 2015, 2017). In a study of two sites on Claridenfirn of the Swiss Alps, Gabbi et al. (2015) showed that mineral dust lowered the mean annual albedo by 0.006–0.011, depending on the location on the glacier, causing a reduction of approximately 142–271 mm in annual mass balance. In the San Juan Mountains of the USA, snow cover duration in a seasonal snow-covered region, was found to be shortened by 18 to 35 days during ablation through surface shortwave RF caused by deposition of disturbed desert dust (Painter et al., 2007). In the upper Colorado River Basin, the daily spring dust RF ranged from 30-65 W m⁻², advancing melt by 15-49 days (Skiles et al., 2015). A study of the Mera glacier on the southern slope of the Himalayas even indicated that, when dust concentrations are high, dust dominates absorption, snow albedo reduction, and RF, and the impact of BC may be negligible (Kaspari et al., 2014). The presence of dust in snow suggests a relevant role for BC in darkening the glacier surface.

References:

- Di Mauro, B., Fava, F., Ferrero, L., Garzonio, R., Baccolo, G., Delmonte, B., Colombo, R.: Mineral dust impact on snow radiative properties in the European Alps combining ground, UAV, and satellite observations, J. Geophys. Res. Atmos., 120, 6080–6097, doi:10.1002/2015JD023287, 2015.
- Di Mauro, B., Baccolo, G., Garzonio, R., Giardino, C., Massabò, D., Piazzalunga, A., Rossini, M., Colombo, R.: Impact of impurities and cryoconite on the optical properties of the Morteratsch glacier (Swiss Alps),

Cryosphere Discuss., doi:10.5194/tc-2017-66, 2017.

Kaspari, S., Painter, T. H., Gysel, M., Skiles, S. M., Schwikowski, M.: Seasonal and elevational variations of black carbon and dust in snow and ice in the Solu-Khumbu, Nepal and estimated radiative forcings, Atmos. Chem. Phys., 14, 8089–8103, 2014.

Line 24: "..., but also to" ! but also important/crucial to Answer: Has been revised. (Page 2, Lin35)

Line 26: ": : : is the most sensitive" how could tell it is the most sensitive? Please remove most or provide supporting data.

Answer: Removed the "most".

Line 33: ": : : and result in perturbation in" ! : : : and perturb Answer: Has been revised. (Page 3, Line 7)

Line 34: "5 – 25 mm in the snow water equivalent: : :"!5-25 mm snow water equivalent Answer: Has been revised. (Page 3, Line 8)

Page 3 Line 1: "snowpack on the TP is associated with the : : :" by associated you mean the snowpack on the TP is influenced by summer monsoon? Or the snowpack will influence summer monsoon? or both? Please be more accurate here.

Answer: Changes in snow cover over the TP have attracted much attention in recent years owing to climate change (Xu et al., 2017). Previous studies have indicated that changes in snow cover across the TP can markedly affect summer monsoons and precipitation over the Indian Ocean (Bai and Feng, 1994; Chen et al., 2000; Vernekar et al., 1995). Through the analysis of spatial and temporal variability of Tibetan snow cover and its relationship with the Indian summer monsoon, Zhao and Moore (2004) showed that there existed an east-west dipole-like correlation pattern between snow cover over the TP and Indian monsoon rainfall that underwent a change in sign around 1985. They also argued that variability in the TP monsoon was responsible for the spatial and temporal variability in the relationship between Tibetan snow cover and the Indian summer monsoon.

The relationship between TP snow cover and the East Asian summer monsoon indicated that winter snow cover played an important role in cooling local air temperature through the snow-albedo effect (Xiao and Duan, 2016). However, data analysis demonstrated that persistent effects of winter snow cover were limited to the period from winter to spring over most parts of the central and eastern TP. Therefore, the preceding snow cover over these regions exerted little influence over either the in situ summer atmospheric heat source or the East Asian summer monsoon, because of its short duration. In contrast, the effects of winter or spring snow cover anomalies over the western TP and the Himalayas can last until summer, and these anomalies further influenced the East Asian

summer monsoon by modulating moisture transport to eastern China and favoring eastwardpropagating synoptic disturbances that were generated over the TP.

Here in the main text has been revised as following (Page3, Line 10-13):

Changes in snow cover over the TP have attracted much attention in recent years owing to climate change (Xiao and Duan, 2016; Xu et al., 2017). Snow cover on the TP plays an important role on the Asian summer monsoon, and serves as a crucial water source for several major rivers of Asia (Bai and Feng, 1994; Lau et al., 2010; Vernekar et al., 1995; Yao et al., 2012a; Zhao and Moore, 2004).

References:

- Bai, Y., and Feng, X.: Introduction to some research work on snow remote sensing, Remote Sens. Technol. Appl., 12(2), 59–65, 1994.
- Chen, Q., Gao, B., Li W., Liu, Y.: Studies on relationships among snow cover winter over the Tibetan Plateau and droughts/floods during Meiyu season in the middle and lower reaches of the Yangtze River as well as atmosphere/ocean. Acta Metall. Sin., 58(5), 582–592, 2000.
- Lau, W. K. M., Kim, M.-K., Kim, K.-M., Lee, W.-S.: Enhanced surface warming and accelerated snow melt in the Himalayas and Tibetan Plateau induced by absorbing aerosols, Environ. Res. Lett., 5, 025204, doi:10.1088/1748-9326/5/2/025204, 2010.
- Vernek, A. D., Zhou, J., Shukla, J.: The effect of Eurasian snow cover on the Indian Monsoon, J. Clim., 8, 248–266, 1995.
- Xiao, Z., and Duan, A.: Impacts of Tibetan Plateau snow cover on the interannual variability of the East Asian summer monsoon, J. Clim., 29, 8495–8514, doi:10.1175/JCLI-D-16-0029.1, 2016.
- Xu, W., Ma, L., Ma, M., Zhang, H., Yuan, W.: Spatial-Temporal variability of snow cover and depth in the Qinghai-Tibetan Plateau, J. Clim., 30, 1521–1533, doi:10.1175/JCLI-D-15-0732.1, 2017.
- Yao, T., Thompson, L.G., Mosbrugger, V., Zhang, F., Ma, Y., Luo, T., Xu, B., Yang, X., Joswiak, D. R., Wang, W., Joswiak, M. E., Devkota, L. P., Tayal, S., Jilani, R., Fayziev, R.: Third Pole Environment (TPE), Environ. Development, 3, 52–64, doi: 10.1016/j.envdev.2012.04.002, 2012a.
- Zhao, H., and Moore, G.W.K.: On the relationship between Tibetan snow cover, the Tibetan Plateau monsoon and the Indian summer monsoon, Geophys. Res. Lett., 31(L14204), doi:10.1029/2004GL020040, 2004.

Line 4-5: "Simulation studies of BC in snow over TP have inherent uncertainties because of the lack of large-area observations of BC data in seasonal snow cover". This is not correct. Model simulations have inherent uncertainties due to the physics/chemistry/transport schemes used in the models - such as uncertainties in BC emissions sources or deposition rate. Large-area observation of BC in TP will be useful for model evaluation, but it will not help with the inherent uncertainty of the model simulation. Please revise it here.

Answer: We agree that large-area observation of BC in TP will be useful for model evaluation. The sentence has been changed to be as following (Page 3, Line15-23):

Simulation studies of BC in snow over the TP have inherent uncertainties due to the physics/chemistry/transport scheme used in the models (Gertler et al., 2016; Yasunari et al., 2010, 2013). For example, Kopacz et al. (2011) estimated RF of 5 to 15 W m⁻² due to BC within the snow-covered areas of Himalaya and the TP, whereas Flanner et al. (2007) and Qian et al. (2011) estimated peak values of BC effects exceeding 20 W m⁻² for some parts of the TP. Menon et al. (2010) and

Ménégoz et al. (2014) proposed that during the last decade BC in snow caused a significant part of the decrease of the snow cover extent or duration observed on the TP during the last decade. Ji et al. (2016b) found a positive surface RF was induced by dust, which caused a decrease of 5–25 mm w.e. over the western TP, Himalayas, and Pamir Mountains from December to May. Large-area observations of BC data in snow cover, which are still seldom in the TP, can be useful for model evaluation and calibration in the future.

Page 4 Line10: "::: and is dominated by the Indian monsoon" ! A region cannot be dominated by monsoon. Do you mean "affected by"?

Answer: It should be "affected by". The sentence has been changed in the main text (Page4, Line27).

Line 11: "Region III has one site (LHG) is located in the northeastern part of the TP"! "Region III has one site (LHG) that is located in the northeastern part of the TP"

Answer: Has changed in the main text as following (Page4, Line28-29).

Intensified sampling has been carried out in the LHG region within Region III, which is located in the northeastern part of the TP.

Line 15: it seems the snow samples were only collected from the top 5 cm, so how did you calculate the snow albedo for snowpacks thicker than 5 cm? Did you assume the BC/MD mass mixing ratio is constant through the entire snow column? Please clarify this.

Answer: We did not measure the BC/MD mass mixing ratio through the entire snow column. Usually, snow samples were collected from the top 5–10 cm. In general, the snow cover is vertically inhomogeneous, and sometimes it is optically thin (Voisin et al., 2012). The stratified structure of snow cover will lead to the discrepancy between computed spectral albedo from semi-infinite snow cover and measured albedo (Grenfell, 1994; Aoki et al., 2000; Zhou et al., 2003; Kuipers Munneke et al., 2008). Recently, the propagation of light in snow with impurities has been extensively investigated with different physical-based numerical models: WW (Grenfell et al., 1994; Jin et al., 2008; Nair et al., 2013), SNICAR (Flanner et al., 2005), PBSAM (Aoki et al., 2000; 2012), GOSWIM (Yasunari et al., 2012; 2014), TARTES (Libios et al., 2013).

In this study, we use SNICAR model to simulate the albedo. This simulator is a single-layer implementation of the Snow, Ice, and Aerosol Radiation model (Flanner et al., 2007, 2009), which utilized the two-stream radiative transfer solution (Toon et al., 1989). Therefore, in this study, we only focused on the surface albedo based on SNICAR model which requests a snow depth of 5 cm. When the snow cover was thicker than 5 cm, the input parameter of "snowpack thickness" (Fig. R1) was the observed snow cover depth.

In the main text (section 2.5), we have added the related information. (Page7, Line24-28):

In general, the snow cover is vertically inhomogeneous, and sometimes it is optically thin (Voisin et al., 2012). The stratified structure of snow cover will lead to the discrepancy between computed spectral albedo from semi-infinite snow cover and measured albedo (Grenfell, 1994; Aoki et al., 2000; Zhou et al., 2003; Kuipers Munneke et al., 2008). In this study, we usually collected snow

samples from the top 5–10 cm. Because the SNICAR model is a single-layer implementation model (Flanner et al., 2007, 2009), the input parameter of "snowpack thickness" was the observed snow cover depth.

SNICAR-Online: Snow albedo simulation

Documentation

la. Incident radiation:
Direct: ®
Diffuse: 0
1b. Solar zenith angle, if incident radiation is direct (0-89 degrees): 60 degrees
2. Surface spectral distribution:
Nid-latitude winter, clear-sky: 🖲
Mid-latitude winter, cloudy:
Summit Greenland, clear-sky: 🔍
Summit Greenland, cloudy: 🛛 🔍
3. Snow grain effective radius (30-1500 microns): 100 $$\mu\rm{m}$$
4. Snowpack thickness: 1 neters
5. Snowpack density: 200 kg/m ³
6. Albedo of underlying ground:
Visible (0.3 - 0.7 µm): 0.2 Near-infrared (0.7 - 5.0 µm): 0.4
7. Black carbon concentration (ppb, or nanograms of BC per gram of ice):
Uncoated: 0 ppb. MAC scaling factor (experimental): 1.0
Sulfate-coated: 0 ppb
8. Dust concentration (ppm, or micrograms of dust per gram of ice):
Size 1 (0.1-1.0 µm diameter): 0 ppm
Size 2 (1.0-2.5 µm diameter): 0 ppm
Size 3 (2.5-5.0 µm diameter): 0 ppm
Size 4 (5.0-10.0 µm diameter): 0 ppm
9. Volcanic ash concentration (ppm, or micrograms of ash per gram of ice): 0 ppm
10. Experimental particle 1 concentration (ppb, or nanograms of particle per gram of ice): 0 ppb
Click "Submit" to display spectral albedo and solar broadband (0.3-5.0 µm) albedo. Submit Reset

Figure R1. The information of SNICAR model input parameters.

References:

- Aoki, T., Aoki, T., Fukabori, M., Hachikubo, A., Tachibana, Y., Nishio, F.: Effects of snow physical parameters on spectral albedo and bidirectional reflectance of snow surface, J. Geophys. Res, 105(D8), 10219–10236, 2000.
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- Grenfell, T. C., Warren, S. G., Mullen, P. C.: reflection of solar radiation by the Antarctic snow surface at ultraviolet, visible, and near-infrared wavelengths, J. Geophys. Res, 99(D9), 18669–18684, 1994.
- Kuipers Munneke, P., Reijmer, C. H., van den Broeke, M. R., König-Langlo, G., Stammes, P., Knpa, W. H.: Analysis of clear-sky Antarctic snow albedo using observations and radiative transfer modelling, J. geophys. Res-Atmos., 113(D17118), doi:10.1029/2007JD009653, 2008.
- Voisin, D., Jaffrezo, J.-L., Houdier, S., Barret, M., Cozic, J., King, M. D., France, J. L., Reay, H. J., Grannas, A., Kos, G., Ariya, P. A., Beine, H. J., Domine, F.: Carbonaceous species and humic like substances (HULIS) in Arctic snowpack during OASIS field campaign in Barrow, J. Geophys. Res., 117(D00R19), doi:10.1029/2011JD016612, 2012.
- Zhou, L., Dickinson, R. E., Tian, Y., Zeng, X., Dai, Y., Yang, Z.-L., Schaaf, C. B., Gao, F., Jin, Y., Strahler, A., Myneni, R. B., Yu, H., Wu, W., Shaikh, M.: Comparison of seasonal and spatial variations of albedos from Moderate-Resolution Imaging Spectradiomater (MODIS) and Common Land Model, J. Geophys. Res, 108(D15), doi:10.1029/JD003326, 2003.

Line 17: how did you measure snow grain size in the field? Please clarify this.

Answer: In the field, the snow grains were sprayed on the MIG paper. Then we took a photo by using portable digital microscope (Anyty 3R-MSV500) (Fig. R2a). In the lab, we can measure the snow grain shape and obtain the lengths of a and b (Fig. R2b). Mugnai and Wiscombe (1987) demonstrated that a collection of unoriented non-spheroids produce the same scattering results as spheres, and Grenfell and Warren (1999) showed that the radius of a non-spherical particle was equal to that of a spherical particle that has the same volume-to-surface-area (V/A) ratio. Consequently, V/A ratio was used to transfer the measured snow grain size into the effective grain size.

On this basis of field measurement, Hao (2009) proposed two assumptions: 1) The snow particle is an inequilateral spheroid; and 2) The major axis, minor axis, and height of the inequilateral spheroid is denoted by a, b, and c, respectively, in such a way that the relationship a = 2b exists. According to Kokhanovsky and Zege (2004), the effective snow grain radius (R_{ef}) can be calculated equal to the radius of the volume-to-surface equivalent sphere by the following equation (1):

$$R_{ef} = \frac{3\bar{V}}{4\bar{A}} \tag{1}$$

Where, $\bar{V} = \frac{4}{3}\pi r^3$ and $\bar{A} = \pi r^2$, are the average volume and the average cross-section (geometric

shadow) area of the snow grains, respectively. And, r is the radius of geometric optics, $r = \frac{a+b}{4}$.

Thus, $R_{ef} \approx 0.35a$

Eq.1 was then employed to calculate the effective snow grain size.

We have added related information in the main text (Page 5, Line2-12):

For the snow grain size observation, we sprayed the snow grains on MIG paper and took a photo using a portable digital microscope (Anyty 3R-MSV500) to calculate the major and minor axis (Fig. S2). Based on field measurements from previous studies (Mugnai and Wiscombe, 1980; Grenfell and Warren, 1999), two assumptions were proposed: 1) The snow grain particle is an inequilateral spheroid, and 2) The major axis (a), minor axis (b), and height (c) of the inequilateral spheroid are denoted by a, b, and c, respectively, in such a way that the relationship a=2b exists (Hao, 2009). According to Kokhanovsky and Zege (2004), the effective snow grain radius (R_{ef}) can be calculated as equal to the radius of the volume-to-surface equivalent sphere by the following equation (1):

$$R_{ef} = \frac{3\bar{V}}{4\bar{A}} \tag{1}$$

where $\bar{V} = \frac{4}{3}\pi r^3$ and $\bar{A} = \pi r^2$, are the average volume and the average cross-section (geometric shadow) area of the snow grains, respectively. In addition, *r* is the radius of geometric optics, $r = \frac{a+b}{4}$.

Thus

$$R_{ef} \approx 0.35a$$
 (2)

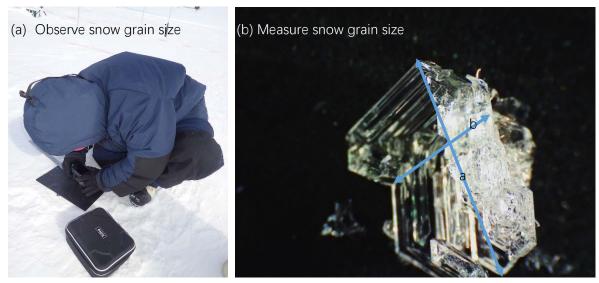


Figure R2. Snow grain size observation in the field (a) and measurement in the lab (b).

References:

Grenfell, T. C., and Warren, S. G.: Representation of a nonspherical ice particle by a collection of independent sphere for scattering and absorption of radiation, J. geophys. Res., 104(D24), 31697–31709, 1999.

Hao, X.: Retrieval of alpine snow cover area and grain size basing on optical remote sensing. PhD thesis, Cold and Arid Regions Environment Engineering Research Institute, Lanzhou, China, pp. 103–104, 2009.

Kokhanovsky, A. A., and Zege, E. P.: Scattering optics of snow, Appl. Optics, 43(7), 1589–1602, 2004.

Mugnai, A., and Wiscombe, W. J.: Scattering of radiation by moderately nonspherical particles. J. Atmos. Sci., 37, 1291–1307, 1987.

Line 24-26: what is the accuracy of the weight you used to measure filter before and after filtration? It seems the author assume BC/OC/MD are the only insoluble particles deposited on the quartz filter, could you please provide more evidence about this? If MD were the only other particle in snow (besides OC and BC), do all MD absorb sunlight? It might be a good idea to include these discussions in the uncertainty analysis.

Answer: The accuracy of the weight for measurement of filter before and after filtration is less than $\pm 5\%$.

We assumed BC/OC/MD are the only insoluble particles deposited on the quartz filter. Not all MD absorb sunlight. Iron oxide minerals are efficient light scattering and absorption materials which can enhance absorption at UV and visible wavelengths (Di Mauro et al., 2015; Moosmuller et al., 2012; Zhang et al., 2015). Goethite and hematite are the most abundant forms of iron oxides in dust and the major light absorbers in the shortwave spectrum in snow (Wu et al., 2016a). The light absorption by mineral dust in snow is thought to be due to iron oxides (Wang et al., 2013). Two peaks in the first derivative value of the spectra at 430 and 560 nm were determined to be goethite and hematite, respectively. When the iron content reaches a threshold, the iron oxides have little or no impact on the reflectance spectra (Wu et al., 2016a). The fine fraction of glacier dust has a greater abundance of iron (2016b), and the first derivative values of hematite are higher than goethite, indicating that hematite might be concentrated in the fine fraction (2016a).

Dust optical properties depend strongly on source material and these properties are designed to represent "global-mean" characteristics as closely as possible (Flanner et al., 2007). The SNICAR model applies the Maxwell-Garnett approximation for combining indices of refractions, assuming a mixture of quartz, limestone, montmorillonite, illite, and hematite. We agree that some dust particles (e.g., those containing a large proportion of strongly-absorbing hematite) can have a larger impact on snow albedo than the dust applied in this work (Aoki et al., 2006; Painter et al., 2007).

We have added related discussion in the section 3.4 as following (Page12, Line25-33):

We also have to pay attention to the fact that dust optical properties depend strongly on source material and their properties are designed to represent "global-mean" characteristics as closely as possible (Flanner et al., 2007). The light absorption by mineral dust in snow is thought to be due to iron oxides (Wang et al., 2013), which are efficient light scattering and absorption materials that can enhance absorption at UV and visible wavelengths (Di Mauro et al., 2015; Moosmüller et al., 2012; Zhang et al., 2015). Goethite and hematite are the most abundant forms of iron oxide in dust and the major light absorbers in the shortwave spectrum in snow (Wu et al., 2016). The SNICAR model applies the Maxwell-Garnett approximation for combining indices of refractions, assuming a mixture of quartz, limestone, montmorillonite, illite, and hematite. We should note that some dust particles (e.g., those containing a large proportion of strongly absorbing hematite) can have a larger impact on snow albedo than the dust applied in this work (Aoki et al., 2006; Painter et al., 2007).

References:

- Aoki, T., Motoyoshi, H., Kodama, Y., Yasunari, T. J., Sugiura, K., Kobayashi, H.: Atmospheric aerosol deposition on snow surfaces and its effect on albedo, SOLA, 2013–2016, doi:10.2151/sola.2006-004, 2006.
- Baccolo, G., Di Mauro, B., Massabò, D., Clemenza, M., Nastasi, M., Delmonte, B., Prata, M., Prati, P., Previtali, E., Maggi, V.: Cryoconite as a temporary sink for anthropogenic species stored in glaciers, Sci. Rep., 7, 9623, doi:10.1038/s41598-017-10220-5, 2017.
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- Moosmüller, H., Engelbrecht, J. P., Skiba, M., Frey, G., Chakrabarty, P. K., Arnott, W. P.: Single scattering albedo of fine mineral dust aerosols controlled by iron concentration, Journal of Geophys. Res., 117(D11210), doi:10.1029/2011JD016909, 2012.
- Painter, T. H., Barrett, A. P., Landry, C. C., Neff, J. C., Cassidy, M. P., Lawrence, C. R., McBride, K. E., Farmer, G. L.: Impact of disturbed desert soils on duration of mountain snow cover, Geophys. Res. Lett., 34, L12502, doi:10.1029/2007GL030284, 2007.
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- Wu, G., Xu, T., Zhang, X., Zhang, C., Yan, N.: The visible spectroscopy of iron oxide minerals in dust particles from ice cores on the Tibetan Plateau, Tellus B, 68, 29191, doi: 10.3402/tellusb.v68.29191, 2016a.
- Wu, G., Zhang, X., Zhang, C., Xu, T.: Mineralogical and morphological properties of individual dust particles in ice cores from the Tibetan Plateau, J. Glaciol., 62(231), 46–53, 2016b.
- Zhang, X., Wu, G., Zhang, C., Xu, T., Zhou, Q.: What is the real role of iron oxides in the optical properties of dust aerosols? Atmos. Chem. Phys., 15, 12159–12177, 2015.

Page 5: Line 6: what is the filter blank for? is this a blank filter? It seems there is some particles on filter blank as well since it weighs more than 0 C/cm-2.

Answer: We used the filter blank to detect the sampling and storage processes. It is a blank filter. The blank filters were brought to the field and stored with the filters together. The results showed that the filter blank is much less $(1.23\pm0.38 \ \mu g \ C \ cm^{-2})$ than those in the sampled filters (more than $10 \ \mu g \ C \ cm^{-2}$), indicating the OC/BC data were reasonable.

Line 8: "separately analyzed" ! analyzed separately Answer: Has been changed.((Page6, Line2)

Line 18: ": : : is or not influenced by BC emissions" ! is influenced by BC emissions or not Answer: Has been changed.((Page6, Line21)

Line 23: what is "down-sun"?

Answer: It's type wrong. Spectral albedo measurements were collected using an ASD FieldSpec 4 (FS4) standard-resolution spectroradiometer in the range of 350–2500 nm using the ASD Remote Cosine Receptor (RCR) foreoptic. The RCR foreoptic was mounted to the end of a 91.4-cm rectangular aluminium boom and levelled at a height of 80 cm above the snow surface using a bubble level located at the observer's end of the boom, collecting sky irradiance when pointed upward and snow irradiance when pointed downward. Spectral albedo was obtained by dividing snow irradiance by sky irradiance from ten consecutive upward and downward measurements collected at site around local solar noon determined using the NOAA SolarNoonCalculator.

We have changed in the main text (Page7, Line2-9).

For the selected sites, a general-purpose spectroradiometer (Analytical Spectral Devices (ASD), FieldSpec 4, Inc.), was used to measure the reflectance of snow cover. The ASD FieldSpec 4 instrument is a general-purpose spectroradiometer that is useful for applications requiring the measurement of reflectance, transmittance, radiance, or irradiance. The instrument is specifically designed for field environment remote sensing to acquire visible and near-infrared and shortwave infrared spectra. It has 3 nm spectral resolution on the visible/near infrared detector (350–1050 nm, silicon photodiode array), and 10–12 nm resolution on the shortwave infrared detectors (900–2 500 nm, InGaAs). In the field, reflectance was measured at two sites (24K and MD in Table S1) with FieldSpec 4 under clear-sky conditions. These measurements of reflectance were calculated using the standard solar irradiance to get the albedos, which were then used for comparison with simulated albedos.

Line 24: "when the weather was clear"! when the sky was clear. It seems the albedo measurement were made for clear sky only, but later in the albedo comparison, the measured albedo is compared

against the albedo calculated using SNICAR for all-sky case. Is this a typo or this is wrong? Answer: Has been changed.

The "all-sky" is a type wrong (see Table S2 in supplementary material), we have changed in the main text.

Line 28: "e.g. Doherty et al., 2010": Doherty et al 2010 did not use SNICAR, it only reported the observation in the Arctic. Please remove this citation. Answer: Has removed.

Page 6: Line 9-11: Please revise this sentence. Is this a model or is this just the method you used to calculate snow water melt in this work?

Answer: It is a method we used to calculate snow water melt. We have changed the sentence in the main text (Page7, Line31-32).

To estimate snow melt due to BC and dust, a method was constructed in which the absorptivity of the snow was multiplied by the daily average incoming shortwave radiation from the automatic weather stations (AWS) set up at the meteorological stations near the sampling sites (Fig. S3) (Schmale et al., 2017).

Line 16: "for clean snow: : :" what is cause of albedo reduction for clean snow case? A different snow grain size due to snow aging? Please be more accurate here.

Answer: Yes. For clean snow, different snow grain size played an important role on the albedo reduction. We have revised the sentence in the main text (Page8, Line7-8):

 $\Delta \alpha$ is the albedo reduction caused by BC and dust, for clean snow mainly by different snow grain sizes due to snow aging;

Line 18: "assumed snow depth" what is the assumed snow depth? How did you assume it? Answer: In the following analysis, we assumed the snow depth to be consistent (which is not the reality) for estimating the variations of snow cover durations days in section 3.3. This assumption was based on the snow depth observations from the TP in previous studies (Che et al., 2012; Xu et al., 2017; Zhong et al., 2016).

We have changed this sentence in the main text (Page8, Line8).

References:

- Che, T., Dai, L., Wang, J., Zhao, K., Liu, Q.: Estimation of snow depth and snow water equivalent distribution using airborne microwave radiometry in the Binggou Watershed, the upper reaches of the Heihe River basin, Int. J. Appl. Earth Obs. Info., 17, 23–32, doi:10.1016/j.jag.2011.10.014, 2012.
- Xu, W., Ma, L., Ma, M., Yuan, W.: Spatial-temporal variability of snow cover and depth in the Qinghai-Tibetan Plateau, J. Clim., doi:10.1175/JCLI-D-150732.1, 2017.
- Zhong, X., Zhang, T., Zheng, L., Hu, Y., Wang, H., Kang, S.: Spatiotemporal variability of snow depth across the Eurasian continent from 1966 to 2012, Cryosphere Discuss., doi:10.5194/tc-2016-182, 2016.

Line 22: "older wind-packed snow" ! old wind-packed snow Answer: Has been changed.(Page8, Line12)

Page 7: Line 2: Please consider cite paper Doherty et al., 2016. It also reports LAIs in snow in North America.

Doherty, S. J., D. A. Hegg, J. E. Johnson, P. K. Quinn, J. P. Schwarz, C. Dang, and S. G. Warren, (2016), Causes of variability in light absorption by particles in snow at sites in Idaho and Utah. J. Geophys. Res. Atmos., 121, no. 9: 4751-4768.

Answer: Agree, and has cited this paper.

Line 6-8: How could you tell the LAIs in snow over TP were generated from biomass burning in surrounding region from Table 2? And from Table 2, how could you tell if the fraction of LAIs emitted by biomass/fossil fuel burning in TP is larger than that in the other regions? Please provide more explanations.

Answer: Table 2 shows the concentrations of LAIs in snow cover in the TP and other regions. These comparisons indicate deposition of LAIs in the snow over the TP have higher values than other regions. In this section, we will not to discuss their fraction and different sources. Thus, we have revised this sentence in the main text as following (Page8, Line27-29):

In comparison (Table 2), the LAIs in snow cover of the TP showed larger values which can be attributed to more deposition from nearby regions around the TP (e.g., South Asia, East Asia, and/or western China) (Lu et al., 2012; Ramanathan and Carmichael, 2008), or to impacts by the soil/dust near the sampling sites.

Line 9-11: How could the ratio of OC/BC be used as a standard to determine the emission sources of biomass burning? What is the OC/BC ratio if all LAIs were emitted from biomass burning or non-biomass burning sources?

Answer: Usually, the aerosols emitted from biomass burning have higher OC/EC ratios. For example, Watson et al. (2001) have reported an OC/EC ratio of 14.5 for forest fires; while for the fossil fuel, OC/EC ratio was about 1 (Watson et al., 2001).

The OC/EC ratios at Mt. Everest station range from 1.91 to 43.8 with an average of 6.69. Such high ratios are considered to be mainly affected by the strong influence of biomass-burning emissions (Cong et al., 2015a), which is also evidenced by the atmospheric organic acids (Cong et al., 2015b).

In the southeastern TP, OC was found to be more abundant during the monsoon season (with OC/EC ratios are 17.67) than in other seasons (e.g. pre-monsoon and winter with OC/EC ratios of 6.29 and 6.45, respectively). These trends can be explained by the emission of plant spores and pollen as well as the formation of greater quantities of secondary organic carbon (SOC) in the periods with the higher OC loadings (Zhao et al., 2013).

We have added related information in the main text as following (Page8, Line30-32):

Ratios of OC to BC (OC/BC) were used to represent the possible impact of biomass burning in previous studies (Watson et al., 2001; Bond et al., 2013; Cong et al., 2015a, 2016b). Usually, the aerosols emitted from biomass burning have higher OC/EC ratios. For example, Watson et al. (2001) reported an OC/EC ratio of 14.5 for forest fires; whereas for fossil fuel, the OC/EC ratio was approximately 1.

References:

- Cong, Z., Kawamura, K., Kang, S., Fu, P.: Penetration of biomass-burning emissions from South Asia through the Himalayas: new insights from organic acids, Sci. Rep., 5, 9580, doi:10.1038/srep09580, 2015b.
- Cong, Z., Kang, S., Kawamura, K., Liu, B., Wan, X., Wang, Z., Gao, S., Fu, P.: Carbonaceous aerosols on the south edge of the Tibetan Plateau: concentrations, seasonality and sources, Atmos. Chem. Phys., 15, 1573-1584, 2015a.
- Watson, J. G., Chow, J. C., Houck, J. E.: PM2.5 chemical source profiles for vehicle exhaust, vegetative burning, geological material, and coal burning in Northwestern Colorado during 1995, Chemos., 43, 1141–1151, 2001.
- Zhao, Z., Cao, J., Shen, Z., Xu, B., Zhu, C., Chen, L.-W. A., Su, X., Liu S., Han, Y., Wang, G., Ho, K.: Aerosol particles at a high-altitude site on the Southeast Tibetan Plateau, China: Implications for pollution transport from South Asia, J. Geophys. Res-Atmos., 118, 11360–11375, 2013.

Line 15: "LHG3 AND LHG6 (Figure 3)", do you mean Figure 1b? Answer: Yes, it should be Figure 1b.

Line 15-21: It seems Figure S2 is an important figure for discussions in this part. If there is no restriction on the number of figures, please consider include this figure in the paper. Answer: Agree, we have included this figure in the main text as Figure 3.

Line 28: "Open burning sourced BC"! BC emitted from open burning sources. Does the BC emitted from open burning sources contain BC emitted from biomass burning? or are they the same? Please be more specific about what you mean by "open burning".

Answer: They are not exactly the same. Here in this study, the results was based on the data of FINN fire emission. Thus, we mean "fire spots or open fire", kind of biomass burning. We have changed this sentence in the main text as following (Page9, Line19-23):

Contributions of BC from open fire burning (a kind of biomass burning) decrease from the southern to the northern TP (Fig. 5). In the Himalayan region, BC from biomass burning sources accounts for half of the BC deposition on snow cover, reflecting the proximity to large sources in South Asia. In the central TP, BC from open fire burning accounts for approximately 30 % of the total, less than from biomass burning evidence from aerosols and glacial snow (Li et al., 2016), maybe indicating a lack of biofuel contributions from our calculation.

Page 8 Line 3-4: "In the southern TP: : :. due to influence of BC sources in the south Asia". This

statement is too vague. Do you mean the total emission is larger in southern Asia? or the BC deposition rate is larger ? or both? This is a major conclusion you have in this work, please be more specific.

Answer: Here we mean the total emission is larger in southern Asia (see from the AOD in Fig. R3) rather than the BC deposition rate. We have changed this sentence in the main text (Page9, Line28-33):

In the southern TP (MYL), the amount of BC deposition is larger than in the northern TP (NETP) due to the nearby BC sources in south Asia, known to be a regional hotspot of BC-induced atmospheric solar heating (Ramanathan and Carmichael, 2008). Whereas in the central TP (NMC), the amount of BC deposition was higher than in the NETP. This may be because pollutants from the southern side of the Himalayas can traverse the high mountain range not only through the major north-south river valleys but also by being lifted and advected over the Himalayas to reach to the inland TP (Cong et al., 2015b; Lüthi et al., 2015).

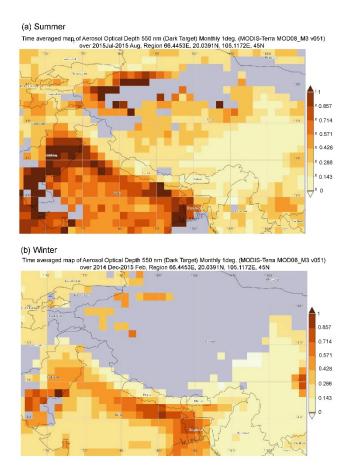


Figure R3. MODIS atmospheric optical depth (AOD) fields derived using dark target algorithms over the TP and its surroundings. (http://giovanni.sci.gsfc.nasa.gov).

Line 8: You used SNICAR model to calculate snow albedo for all-sky cases, but the model set up is not stated. For example, what is the cloud fraction for all sky case? As mentioned in the previous comments, the albedo measurement was performed under clear sky, so in what accuracy do you expect this to agree with the SNICAR calculation (Figure 5)?

Answer: Here is not the all-sky cases, it should be clear sky. We have changed in the main text.

Line 14: "The deviation between measured and simulated reflectance by MD may be a result of the upper boundary of the SNICAR model in particle dimension". This does not make sense, is the deviation due to upper boundary condition (and what upper boundary condition)? or MD particle size? Please revise.

Answer: The difference between measured and simulated reflectance by MD may be caused by the upper boundary of particle size (as shown in Fig. R1, the dust concentrations) in the SNICAR model (Flanner et al., 2007).

Thus, we have changed this sentence in the main text (Page10, Lin10-11).

Line 18-19: "This result is import, showing that the SNICAR model simulations can represent albedo changes of snow cover in the Third Pole region". This is a really strong conclusion. I don't think the authors can make this conclusion based on the results shown in this paper. Especially it is unclear to the readers that how did the authors set up the SNICAR calculation. Please remove this or include more details about SNICAR calculation.

Answer: Agree, and removed this sentence.

Line 26. As the author said, the BC snow albedo forcing over TP is highly uncertain partly due to the uncertainty in simulated BC concentration. But it is also important to point out that a large fraction of such uncertainty is resulted form uncertainty in simulated snow-cover fraction/snow depth. Please consider including this in the discussion.

Answer: Agree, we have added this section in the discussion (Page13, Line1-10).

It is further important to note that a large fraction of such uncertainty can also result from uncertainty in the simulated snow-cover fraction/snow depth across the plateau. The TP covers a large area (more than 2.5 million km²) with an average elevation exceeding 4000 m a.s.l. (Yao et al., 2012a). Considerable heterogeneity in the topography and climate has led to complex spatial and temporal snow cover patterns (Xu et al., 2017). Most studies on the snow cover distribution were based on satellite-based observations (Che et al., 2008, 2012). Surface observations of snow depth showed an increase over the TP from 1957–1998 (Ma and Qin, 2012). However, snow cover depth and the number of snow covered days during the current decade under intense climate warming showed a decreasing trend mainly occurring in the southeast TP in winter. Whereas in spring, snow cover depth showed an increasing trend in the eastern TP (Xu et al., 2017). These differences can also affect the estimation in this study. Nevertheless, the method provides a theoretical approach for evaluating how the presence of LAIs affects the lower parts of the glacier subjected to summer melt.

References:

- Che, T., Dai, L., Wang, J., Zhao, K., Liu, Q.: Estimation of snow depth and snow water equivalent distribution using airborne microwave radiometry in the Binggou Watershed, the uppwe reaches of the Heihe River basin, Int. J. Appl. Earth Obs. Info., 17, 23–32, doi:10.1016/j.jag.2011.10.014, 2012.
- Che, T., Li, X., Jin, R., Armstrong, R., Zhang, T.: Snow depth derived from passive microwave remote-sensing data

in China, Ann. Glaciol., 49, 145-154, doi:10.3189/172756408787814690, 2008.

- Ma, L., Qin, D.: Temporal–spatial characteristics of observed key parameters of snow cover in China during 1957– 2009, Sci. Cold Arid Reg., 4, 384–393, doi:10.3724/SP.J.1226.2012.00384, 2012.
- Xu, W., Ma, L., Ma, M., Zhang, H., Yuan, W.: Spatial-Temperal variability of snow cover and depth in the Qinghai-Tibetan Plateau, J. Clim., 30, 1521–1533, doi:10.1175/JCLI-D-15-0732.1, 2017.
- Yao, T., Thompson, L.G., Mosbrugger, V., Zhang, F., Ma, Y., Luo, T., Xu, B., Yang, X., Joswiak, D. R., Wang, W., Joswiak, M. E., Devkota, L. P., Tayal, S., Jilani, R., Fayziev, R.: Third Pole Environment (TPE), Environ. Development, 3, 52–64, doi: 10.1016/j.envdev.2012.04.002, 2012a.

Page 9 Section 3.3: Why did you pick SW flux of 220, 270 and 310 W m-2?

Answer: We selected short-wave radiation from several automatic weather stations (Fig. R4) near the snow sampling sites in region I, II, and III. Table R1 showed monthly short-wave radiation from automatic weather stations near the snow sampling sites during the snow melting season (March-May) when the temperature began to increase. On average, shortwave radiations in March, April, May, and June is about 238, 269, 292, and 271 W m⁻², respectively. Short-wave radiation showed the minimum value in March in Tanggula (210 W m⁻², lower than the average) and the maximum value in May in Namco (314 W m⁻², higher the average). Thus, in order to estimate the impact of input short-wave radiation on the snow cover duration, we gave a range of short-wave radiation from 220 to 310 W m⁻².

We have added related information in the main text as following (Page11, Line14-23): Changes of snow cover duration are calculated using a model documented by Schmale et al. (2017) (section 2.6). In Eq. (4), monthly shortwave radiation (*SW*) input data were obtained from the Automatic Weather Station (AWS) near the snow sampling sites across the TP (Fig. S3). Table S3 shows monthly shortwave radiation data from AWS during the snow melting season (March-May) when the temperature began to increase. On average, shortwave radiation in March, April, May, and June is approximately 238, 269, 292, and 271 W m⁻², respectively. Short-wave radiation in March showed the minimum value in Tanggula of the central TP (210 W m⁻², lower than the average), whereas in May it showed the maximum value in Namco (314 W m⁻², higher the average). Based on these data during the melt season, three scenarios (220, 270, and 310 W m⁻²)were defined as the minimum, median, and maximum scenarios of input shortwave radiation to estimate its impact on changes of snow cover duration.

We have added these information in the supplementary material as Table S3.

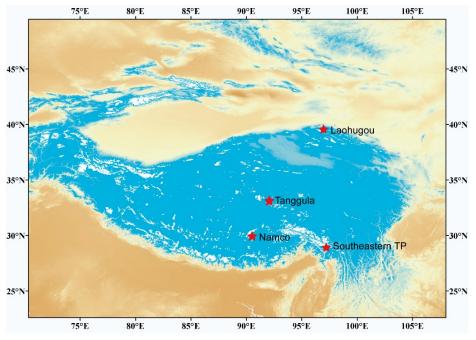


Figure R4. Automatic weather stations selected in the Tibetan Plateau.

Sites	Month of the year	Monthly Short-wave radiation (W m ⁻²)	Month of the year	Monthly Shortwave radiation (W m ⁻²)
Southeastern				
Tibetan Plateau				
	March in 2014	248	March in 2015	237
	April in 2014	288	April in 2015	259
	May in 2014	305	May in 2015	280
	June in 2014	265	June in 2015	250
Namco				
			March in 2015	264
			April in 2015	276
			May in 2015	306
			June in 2015	314
Tanggula				
	March in 2014	210		
	April in2014	226		
	May in 2014	245		
	June in 2014	271		
Laohugou				
	March in 2014	229	March in 2015	238
	April in2014	269	April in 2015	294
	May in 2014	311	May in 2015	305
	June in 2014	258	June in 2015	269

Table R1. Monthly short-wave radiation from automatic weather stations near the snow sampling sites during the snow melting season.

Line 27: "SD plays an import role" ! SD plays an important role? What do you mean by SD plays a role here? Deeper snow is supposed to melt slower given the same amount of radiative forcing; it is not because snow depths play a role here. Please clarify this point.

Answer: We agree that deeper snow is supposed to melt slower given the same amount of radiative forcing. In this study, we calculated the changes of snow cover duration days rather than the total days of snow cover duration. We mean that SD plays an important role on our estimation of numbers of snow cover duration days. Thus, we have changed the sentence in the main text as following (Page11, Line28-29).

The result indicated that estimation of changes of snow cover duration days caused by the same level of LAIs was also affected by different SD.

Page 10: Line 13: "However, the results presented in this study : : :. for which these assumptions are not critical": This is not true. All the quantities listed in this paragraph will influence the snow albedo and most of them will influence the albedo reduction induced by BC. For example: BC-snow internal mixing increases the albedo forcing by 40-60% compared with external mixing (He et al. (2014). The author should discuss the uncertainty of this study resulted from the assumptions they made, instead of claim these quantities will not impact their results.

References: He, C., Q. Li, K. N. Liou, Y. Takano, Y. Gu, L. Qi, Y. Mao, and L. R. Leung, 2014: Black Carbon Radiative Forcing over the Tibetan Plateau. Geophy. Res. Lett., 41, 7806-7813, doi: 10.1002/2014GL062191.

Answer: Agree, and we have added related discussion in this section in the main text as suggested.

Line 32: "Our study confirms that : : ... and further reduces snow albedo,: : :..": this is not true. BC and other LAI can reduce the snow albedo even if the snow aging process is not accelerated. Please revise this.

Answer: Agree, and have changed (Page13, Line26).

Our study confirms that BC and other water-insoluble LAIs in snow on land and ice can darken the surface, and further reduces snow albedo, and increases the speed of snow cover melt

Figures3: Is the color bar showing height? Please define the color bar and unit. Answer: Yes, the color bar shows the height. We have added the information in the figure caption.

Figure 4: Footprint analyses for six selected sites over the Tibetan Plateau during the winter season (Nov 2015-Feb 2016). LHG and NETP in the northern Tibetan Plateau (Region III), TGL and NMC in the central Tibetan Plateau (Region II), and MYL and SETP in the southern Tibetan Plateau (Region I). (The right color bar means the height (m a.s.l.). The trajectories starting below 500 hPa were taken into account. Black dots: the air parcel did not pass near a fire during the 96 h prior to arrival at the studied site; Green dots: the air parcel did pass by a fire between -96 h and -48 h, but not afterwards, i.e., the 'contact' to a fire lies back at least 48 h before the air parcel arrived at Renlongba glacier; Magenta dots: contact with fires occurred between -48 h and 0 h before arriving at site; Red dots: contact with a fire before and after -48 h.)

Figure 5: Are MA1-4 measured albedo? Are the dashed lines albedo calculated using SNICAR? Please clarify these details and modify the corresponding text.

Answer: In this figure, MA1-4 (solid lines) are measured albedo. Dashed lines mean the albedo calculated using SNICAR. We have added the information in the figure caption.

Figure 6: Measured albedo by ASD (solid lines, MA1-4) and simulated effects of BC and dust on albedo (dashed lines) at the selected snow site on the Tibetan Plateau.

Figure 6-8: please clarify the figure convention in each figure. Do the boxes represent average values from central estimate? For example, in Figure 6, you say the rectangles are central estimate, so what does the box mean? standard deviation of central estimate? maximum and minimum of central estimate?

Answer: We have added related information and legend for each figure in the main text.

Oct10, 2017

Dear editor,

The manuscript: *Black carbon and mineral dust in snow cover on the Tibetan Plateau* by Zhang Y.L. et al.

The manuscript number: tc-2017-111

We greatly appreciate the reviewers' constructive comments to improve the paper. We have revised our manuscript according to these comments (blue color in the main text), and hope the revised manuscript is suitable for publication in The Cryosphere.

The "point to point" response to comments are listed as below.

Sincerely yours, Yulan Zhang and Shichang Kang

Response to Referee #2

This manuscript presents results from snow sampling, laboratory analysis, and related modeling efforts for the presence and impact of light absorbing impurities in the Tibetan Plateau region. Although the manuscript is not wrong in stating more measurements are valuable in constraining our understanding of the spatial and temporal variability of light absorbing particulates, the techniques presented in this paper are unclear and as written does not contribute to the state of knowledge of light absorbing particulates in snow. Major changes are needed before this paper can be reviewed again for publication. I will not, at the point, do line by line corrections because they are numerous. The authors need to revisit the writing in each section for editing and to clarify there justification, methods, and results. Particularly, snow sampling and automatic weather stations need to be described in significantly more detail.

Answer: Thank you very much for the comments and suggestions. We have tried to improve the methods and results in the main text (in blue color). Snow sampling and automatic weather stations are also described in detailed (Section2.2, Section3.3, in blue color).

Comment 1: My first and foremost concern is that samples were collected in November and December, and yet they are attempting to quantify the impacts of light absorbing particulates on melt. This does not make sense to me and if I misunderstood this it is because it is not made clear in the manuscript. Although the sample collection timing may be after the summer monsoon, these samples do not represent the impurities that are present during the ablation season- and therefore it is inappropriate to use these values to quantify reduction in snowpack duration. Particularly for dust, which tends to deposit in the spring when source regions dry out (peak radiative forcing by dust in snow is observed from MODIS imagery over the Himalayan region in April and May).

Answer: Agree, these samples were collected during winter season when the snow cover were more stable and continuous, which might not represent the true impurities as these during the ablation season. However, during melting season, because of the poor accessibility on the snowpack in the TP, it was hardly to collect the snow cover samples. Besides, considerable heterogeneity in the topography and climate has led to complex spatial and temporal snow cover patterns (Xu et al., 2017). Discontinued snow cover during melting season may be a problem to represent the true impurities in snow. Thus in our study, we gave an estimation based on the different snow grain size and density, rather than the fixed data. We have to admit there existed uncertainties at present. In the future, we should do more related works to fill in gaps.

Comment 2: The backtrajectory footprint modeling was also very unclear to me, how the model runs were carried out needs to be described better, but it is unclear to me why the model runs were only completed for the winter. And were they run continuously? Typically particulates are deposited in episodic events so running it continuously does not inform the source region, it just informs of the regional synoptics. I suggest seeing Skiles et al., 2015 (Hydrological Processes) for how that study produced and described backtrajectory footprints.

Answer: We agree. Since most of the snow cover will not exist during summer season in the TP, thus we only calculated the model runs in winter when the snow cover accumulated. The calculation is continuous in the winter season since we did not know the exact snow events during the periods. It is an estimation of all air mass which can be transported to the sampling sites.

We have learned the recommended reference (Skiles et al., 2015), and tried to improve the descriptions in the main text as following (Page6, Line6-34).

Calculation of air parcel trajectories is a widely used approach in different areas of atmospheric research. Conceptually, footprint analysis was considered as changes in concentration at the receptor site that can be attributed to different upwind source areas along the backward trajectories (Skiles et al., 2015). To determine the potential origins of the LAIs deposited on the snow cover of the TP, back trajectory analyses were performed using the European Centre for Medium-Range Weather Forecasts (ECMWF) analysis fields with the Lagrangian analysis tool LAGRANTO (Sprenger and Wernli, 2015), launched every six hours for six selected sampling sites as receptors (including three sites of MYL, NMC, and SETP in region I, two sites of TGL and NETP in region II, and LHG in region III) during the winter season. The ECMWF fields (horizontal and vertical wind components) were retrieved on 137 model levels and then interpolated onto a $0.25^{\circ} \times 0.25^{\circ}$ latitude-longitude grid. Trajectory starting positions can be defined easily and flexibly based on different geometrical and meteorological conditions; after the computation of the trajectories, a versatile selection is offered based on single or combined criteria (Sprenger and Wernli, 2015). First, starting positions are initialized with a suitable domain over the TP at 12:00 UTC on November 1, 2015 (or 2014). For this case, a domain from 0 to 75 and 60 $^{\circ}$ E to 120 $^{\circ}$ E was chosen. We choose starting positions in this domain that are horizontally equidistant with 80 km horizontal spacing and extend vertically from 1030 to 790 hPa with 30 hPa vertical spacing. Then, the trajectories are calculated from all starting positions 96 h backward in time. Finally, biomass burning emission data is traced along the calculated trajectories to estimate whether an air parcel at the receptor site is influenced by BC fire emissions or not. In this study, the Fire INventory from NCAR (FINN) v1.5 global fire emissions flux in 2012–2014, speciated with the GEOS-chem mechanism, was used to estimate contributions of BC fire emission at the six selected receptor sites. FINN emission estimates are based on the framework described by Wiedinmyer et al. (2011). FINN used the satellite observations of active fires and land cover, together with emission factors and estimated fuel loadings to provide daily, highly resolved (1 km) open biomass burning emissions estimates for use in regional and global chemical transport models. Then, the same calculation was performed for the Eclipse V5 inventory for the anthropogenic contributions of BC. The Eclipse V5 inventory was widely used in the simulations. The historical data for the period 1990–2010 were revised compared to preceding sets using the latest IAE (the International Energy Agency) and FAO (the Food and Agriculture Organization) statistics extending to 2010, as well as recent country reporting where available. Note that this analysis was not exactly quantitative and did not take into account wet and dry deposition. We also assumed that the anthropogenic emissions have not changed significantly from 2010 compared to the period from 2012–2014. Thus, the comparison of natural and anthropogenic BC contributions was reasonable. The relative differences between the BC contributions can provide information on regional differences in this study. References:

Skiles, S. M., Painter, T. H., belnap, J., Holland, L., Reynolds, R. L., Goldstein, H. L., Lin, J.: Regional variability in dust-on-snow processes and impacts in the Upper Colorado River Basin, Hydrol. Process., doi:10.1002/hyp.10569, 2015.

Comment 3: The albedo measurements need to be better described. And how was snow effective grain radius retrieved? This should be an optically equivalent grain size, not an observable grain size. If an effective/optical grain size was used, the retrieval should be described. If observed grain sizes were used, the large error introduced by this (see Painter et al., 2007 in Journal of Glaciology) needs to be mentioned. Estimates of changes in snow cover duration should be removed or significant more detail and justification needs to be made for the timing of sample collection. Answer: In this study, we used an optically equivalent grain size, not an observable grain size.

In the field, the snow grains were sprayed on the MIG paper. Then we took a photo by using portable digital microscope (Anyty 3R-MSV500) (Fig. R1a). In the lab, we can measure the snow grain shape and obtain the lengths of a and b (Fig. R1b). Mugnai and Wiscombe (1987) demonstrated that a collection of unoriented non-spheroids produce the same scattering results as spheres, and Grenfell and Warren (1999) showed that the radius of a non-spherical particle was equal to that of a spherical particle that has the same volume-to-surface-area (V/A) ratio. Consequently, V/A ratio was used to transfer the measured snow grain size into the effective grain size.

On this basis of field measurement, Hao (2009) proposed two assumptions: 1) The snow particle is an inequilateral spheroid; and 2) The major axis, minor axis, and height of the inequilateral spheroid is denoted by a, b, and c, respectively, in such a way that the relationship a = 2b exists. According to Kokhanovsky and Zege (2004), the effective snow grain radius (R_{ef}) can be calculated equal to the radius of the volume-to-surface equivalent sphere by the following equation (1):

$$R_{ef} = \frac{3\bar{\nu}}{4\bar{A}} \tag{1}$$

Where, $\overline{V} = \frac{4}{3}\pi r^3$ and $\overline{A} = \pi r^2$, are the average volume and the average cross-section (geometric

shadow) area of the snow grains, respectively. And, r is the radius of geometric optics, $r = \frac{a+b}{4}$.

Thus, $R_{ef} \approx 0.35a$

Eq.1 was then employed to calculate the effective snow grain size.

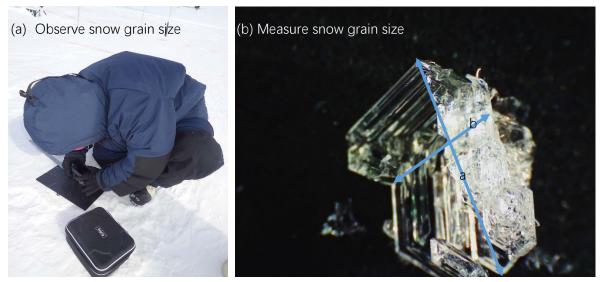
We have added related information in the main text (Page 5, Line2-12):

For the snow grain size observation, we sprayed the snow grains on MIG paper and took a photo using a portable digital microscope (Anyty 3R-MSV500) to calculate the major and minor axis (Fig. S2). Based on field measurements from previous studies (Mugnai and Wiscombe, 1980; Grenfell and Warren, 1999), two assumptions were proposed: 1) The snow grain particle is an inequilateral spheroid, and 2) The major axis (a), minor axis (b), and height (c) of the inequilateral spheroid are denoted by a, b, and c, respectively, in such a way that the relationship a=2b exists (Hao, 2009). According to Kokhanovsky and Zege (2004), the effective snow grain radius (R_{ef}) can be calculated as equal to the radius of the volume-to-surface equivalent sphere by the following equation (1):

$$R_{ef} = \frac{3\overline{\nu}}{4\overline{A}} \tag{1}$$

where $\bar{V} = \frac{4}{3}\pi r^3$ and $\bar{A} = \pi r^2$, are the average volume and the average cross-section (geometric shadow) area of the snow grains, respectively. In addition, *r* is the radius of geometric optics, $r = \frac{a+b}{4}$.

Thus $R_{ef} \approx 0.35a$



(2)

Figure R1. Snow grain size observation in the field (a) and measurement in the lab (b).

For the estimates of changes in snow cover duration days, we have tried to improve the related details in the main text.

References:

- Grenfell, T. C., and Warren, S. G.: Representation of a nonspherical ice particle by a collection of independent sphere for scattering and obsorption of radiation, J. geophys. Res., 104(D24), 31697–31709, 1999.
- Hao, X.: Retrieval of alpine snow cover area and grain size basing on optical remote sensing. PhD thesis, Cold and Arid Regions Environment Engineering Research Institute, Lanzhou, China, pp. 103–104, 2009.
- Mugnai, A., and Wiscombe, W. J.: Scattering of radiation by moderately nonspherical particles. J. Atmos. Sci., 37, 1291–1307, 1987.

Comment 4: Section 3.3 and 3.4 are generally confusing- was shortwave radiation (uplooking and downlooking pyranometers) actually measured? This analysis seems far too simplified. Furthermore, the discussion of snow depth is misleading. This is a straightforward energy balance calculation, so less snow will melt faster than more snow for an equal amount of forcing- this is basic mass balance. So snow depth does not itself play an important role in the reduction of snow cover by particulates. This is a mixing up of forcing and state functions. The citations are also outdated or incorrect in many cases, and I suggest the authors revisit these.

Answer: Shortwave radiation data used in this study was actually measurement by automatic weather station at different selected site near the snow sampling sites in region I, II, and III of the TP (Fig R2). Table R1 showed monthly short-wave radiation from automatic weather stations near the snow sampling sites during the snow melting season (March-May) when the temperature began to increase. On average, shortwave radiations in March, April, May, and June is about 238, 269, 292, and 271 W m⁻², respectively. Short-wave radiation in March showed the minimum value in Tanggula (210 W m⁻², lower than the average). Short-wave radiation in May showed the maximum value in Namco (314 W m⁻², higher the average). Thus, in order to estimate the impact of input short-wave radiation on the snow cover duration, we gave a range of short-wave radiation from 220 to 310 W m⁻².

We have added related information in the main text as following (Page11, Line14-23): In Eq. (4), monthly shortwave radiation (*SW*) input data were obtained from the Automatic Weather Station (AWS) near the snow sampling sites across the TP (Fig. S3). Table S3 shows monthly shortwave radiation data from AWS during the snow melting season (March-May) when the temperature began to increase. On average, shortwave radiation in March, April, May, and June is approximately 238, 269, 292, and 271 W m⁻², respectively. Short-wave radiation in March showed the minimum value in Tanggula of the central TP (210 W m⁻², lower than the average), whereas in May it showed the maximum value in Namco (314 W m⁻², higher the average). Based on these data during the melt season, three scenarios (220, 270, and 310 W m⁻²)were defined as the minimum, median, and maximum scenarios of input shortwave radiation to estimate its impact on changes of snow cover duration.

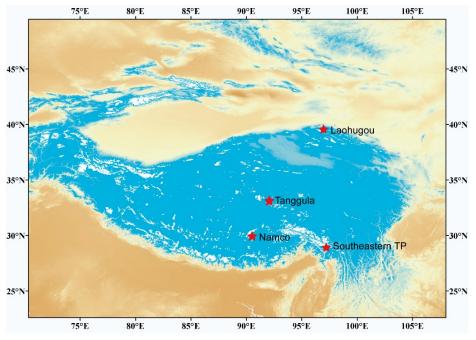


Figure R4. Automatic weather stations selected in the Tibetan Plateau.

Sites	Month of the year	Monthly Short-wave radiation (W m ⁻²)	Month of the year	Monthly Shortwave radiation (W m ⁻²)
Southeastern				
Tibetan Plateau				
	March in 2014	248	March in 2015	237
	April in 2014	288	April in 2015	259
	May in 2014	305	May in 2015	280
	June in 2014	265	June in 2015	250
Namco				
			March in 2015	264
			April in 2015	276
			May in 2015	306
			June in 2015	314
Tanggula				
	March in 2014	210		
	April in2014	226		
	May in 2014	245		
	June in 2014	271		
Laohugou				
	March in 2014	229	March in 2015	238
	April in2014	269	April in 2015	294
	May in 2014	311	May in 2015	305
	June in 2014	258	June in 2015	269

Table R1. Monthly short-wave radiation from automatic weather stations near the snow sampling sites during the snow melting season.

We agree that the equation for changes of snow cover duration is a straightforward energy balance calculation. Thus in this study, it means the duration days of less snow will be less than the more snow for an equal amount of forcing. So under the same level of LAIs in snow, the reduction of snow cover duration days can be affected by different snow depth. We have tried to revise in the main text (section 3.4).

Comment 5: I take issue with the use of the 'Third Pole' term, this is not universally recognized and is somewhat politicized, why not just use Tibetan Plateau or High Mountain Asia? Also light absorbing impurity is an outdated term, the community has moved toward the use of light absorbing particulates. Also mineral dust and the acronym MD are confusing you can simply say dust-which needs no acronym. Similarly, please be consistent in terminology, for example, albedo and reflectance are not the same thing.

Answer: We have changed the term "Third Pole" to be "Tibetan Plateau".

We have used the term "light-absorbing particulates" in the main text.

We have changed the "MD" as "dust" in the main text.

Albedo and reflectance are not the same thing. Albedo is defined as the ratio of irradiance reflected to the irradiance received by a surface. The proportion reflected is not only determined by properties of the surface itself, but also by the spectral and angular distribution of solar radiation reaching the Earth's surface. Unless given for a specific wavelength (spectral albedo), albedo refers to the broadband spectrum of solar radiation.

Reflectance of the surface of a material is its effectiveness in reflecting radiant energy. It is the fraction of incident electromagnetic power that is reflected at an interface. The reflectance spectrum or spectral reflectance curve is the plot of the reflectance as a function of wavelength. When we use ASD, we get the reflectance. In order to compare the albedo simulated by SNICAR model in this study, we have to calculate the reflectance based on the standard solar irradiance. We also carefully use the words consistent in the main text.

Comment 6: Uncertainties in modeling are not only due to lack of observations, and increasing our number of sampling points alone will not reduce our model uncertainty. To state this is misleading. Answer: Agree, and we have tried to revise this section in the section 3.4 of the main text.

Black carbon and mineral dust in snow cover on the Tibetan Plateau

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Abstract. Snow cover plays a key role for sustaining ecology and society in mountainous regions. Light-absorbing particulates (including black carbon, organic carbon, and mineral dust) deposited on snow can reduce surface albedo and

- 15 contribute to the near-worldwide melting of snow cover and ice. This study focused on understanding the role of black carbon and other water-insoluble light-absorbing particulates in the snow cover of the Tibetan Plateau (TP). The results found that the black carbon, organic carbon, and dust concentrations in snow cover generally ranged from 202–17 468 ng g⁻¹, 491–13 880 ng g⁻¹, and 22–846 μ g g⁻¹, respectively, with higher concentrations in the central to northern areas of the TP. Footprint analyses suggested that the northern TP was influenced mainly by air masses from Central Asia with some Euro-
- 20 Asia influence, and air masses in the central and Himalayan region originated mainly from Central and South Asia. The biomass burning sourced black carbon contributions decreased from ~50 % in the southern TP to ~30 % in the northern TP. The contribution of black carbon and dust to snow albedo reduction reached approximately 37 % and 15 %, respectively. The effect of black carbon and dust reduced the average changes of snow cover duration by 3.1±0.1 days to 4.4±0.2 days. Meanwhile, the black carbon and dust had important implications for snowmelt water loss over the TP. The findings indicate
- that the impacts of black carbon and mineral dust need to be properly accounted for in future regional climate projections, particularly in the high-altitude cryosphere.

1 Introduction

5

Black carbon (BC), organic carbon (OC), and mineral dust (hereafter: dust) are the main constituents of light-absorbing
particulates (LAPs) in snow (Andreae and Gelencsér, 2006; Bond et al., 2013; Di Mauro et al., 2015; Doherty et al., 2010;
Painter et al., 2010). LAPs can reduce snow and ice albedo and trigger positive feedbacks that contribute to the near-worldwide melting of snow cover and ice (Dumont et al., 2014; Flanner et al., 2007; Hadley and Kirchstetter, 2012; Hansen

and Nazarenko, 2004; Painter et al., 2013; Zhang et al., 2017). Atmospheric BC is a distinct type of carbonaceous material from incomplete combustion of biomass/biofuel and fossil fuel. A large fraction of BC is emitted from anthropogenic activities (Bond et al., 2013). Because BC can absorb solar radiation, influence cloud processes, and alter the melting of snow cover and glaciers, it has been considered to be the second most important climate forcer in the Earth's climate system

- 5 only after carbon dioxide (Andreae and Ramanathan, 2013; Bond et al., 2013; Ramanathan and Carmichael, 2008). Model studies in recent decades indicated that BC in seasonal snow may be responsible for a significant impact on warming and albedo reduction over the Arctic (Flanner, 2013; Qian et al., 2014), Greenland (Dumont et al., 2014), the Alps (Painter et al., 2013; Yasunari et al., 2015), and the Tibetan Plateau (TP) (He et al., 2014; Jacobi et al., 2015; Ji et al., 2015; Ménégoz et al., 2014). The snow albedo feedback of BC may also be an important driver accelerating glacial retreat and snow cover melt
- (Painter et al., 2013; Skiles et al., 2015; Xu et al., 2009).
 Light-absorbing OC in snow can contribute significantly to the absorption by ultraviolet/visible light (Andreae and Gelencsér, 2006; Yan et al., 2016). Bahadur et al. (2012) studied aerosol optical properties derived from the Aerosol Robotic Network (AERONET) measurements and estimated that, in California, the OC absorption at 440 nm was approximately 40 % of the BC absorption. Measurements of hundreds of snow samples revealed that approximately 40 % of the light
- 15 absorption from impurities in Arctic snow and sea ice is due to non-BC constituents (Doherty et al., 2010). In western North America, Dang and Hegg (2014) suggested that OC was responsible for more than 10 % of the total light absorption. A global model by Lin et al. (2014) estimated the role of OC in the reduction of snow albedo and the simulation showed that the radiative forcing (RF) of OC deposited in land snow and sea ice ranges from +0.0011 to +0.0031 W m⁻² or as large as 24 % of the forcing caused by BC in snow and ice. The RF of snow pit dissolved organic carbon (DOC) from a Tibetan glacier
- 20 was calculated to be 0.43 W m^{-2} , indicating that DOC in snow needs to be taken into consideration in accelerating glacial melt on the TP (Yan et al., 2016).

Mineral dust, another important LAP component in snow cover, can change the cryospheric environment and hydrological cycle due to its light-absorbing properties (Di Mauro et al., 2015; Ji et al., 2016a; Painter et al., 2010, 2013; Wu et al., 2016; Zhang et al., 2015). In the European Alps, observed reflectance provided evidence that seasonal input of dust can strongly

- 25 decrease the spectral properties of snow, in particular, from 350 to 600 nm (Di Mauro et al., 2015, 2017). In a study of two sites on Claridenfirn of the Swiss Alps, Gabbi et al. (2015) showed that mineral dust lowered the mean annual albedo by 0.006–0.011, depending on the location on the glacier, causing a reduction of approximately 142–271 mm in annual mass balance. In the San Juan Mountains of the USA, snow cover duration in a seasonal snow-covered region, was found to be shortened by 18 to 35 days during ablation through surface shortwave RF caused by deposition of disturbed desert dust
- 30 (Painter et al., 2007). In the upper Colorado River Basin, the daily spring dust RF ranged from 30–65 W m⁻², advancing melt by 15-49 days (Skiles et al., 2015). A study of the Mera glacier on the southern slope of the Himalayas even indicated that, when dust concentrations are high, dust dominates absorption, snow albedo reduction, and RF, and the impact of BC may be negligible (Kaspari et al., 2014). The presence of dust in snow suggests a relevant role for BC in darkening the glacier surface.
 - 2

As an important component of the hydrology, snow cover plays a key role for sustaining ecology and society in mountainous regions (Beniston et al., 2017). Snowpack changes are not only crucial for estimating impacts on water resources but also important to anticipate natural hazards related to snow avalanches/disasters (Beniston et al., 2017; Brown and Mote, 2009). Covered by a large volume of snow/ice in the mid-low latitudes, the TP is considered to be a sensitive and readily visible

- 5 indicator of climate change (Yao et al., 2012a). Recently, the TP cryosphere has undergone rapid changes (Kang et al., 2010), including glacial shrinkage (Xu et al., 2009; Yao et al., 2012b), permafrost degradation (Cheng and Wu, 2007), and reduction in the annual duration of snow cover days (Ménégoz et al., 2014; Xu et al., 2017). Studies on the TP indicate that BC has been a significant contributing factor to the observed cryospheric change (Li et al., 2017; Ming et al., 2009; Niu et al., 2017; Xu et al., 2009;Yang et al., 2015; Zhang et al., 2017), and a major forcer of climate change (Qian et al., 2015; Ji, Ji, Ji)
- 10 2016a). Observations and simulations also found that dust deposition on snow/ice can change the surface albedo and perturb the surface radiation balance (Ji et al., 2016b; Qu et al., 2014), causing a decrease of 5–25 mm snow water equivalent (mm w.e.) over the western TP and Himalayas (Ji et al., 2016b).

Changes in snow cover over the TP have attracted much attention in recent years owing to climate change (Xiao and Duan, 2016; Xu et al., 2017). Snow cover on the TP plays an important role in the Asian summer monsoon, and serves as a crucial

- 15 water source for several major Asian rivers (Bai and Feng, 1994; Lau et al., 2010; Vernekar et al., 1995; Yao et al., 2012a; Zhao and Moore, 2004). Atmospheric heating related to BC and dust deposition can lead to widespread enhanced warming over the TP and accelerated snowmelt in the western TP and Himalayas (Lau et al., 2010; Ramanathan and Carmichael, 2008; Zhang et al., 2015). Simulation studies of BC in snow over the TP have inherent uncertainties due to the physics/chemistry/transport scheme used in the models (Gertler et al., 2016; Yasunari et al., 2010, 2013). For example,
- 20 Kopacz et al. (2011) estimated RF of 5 to 15 W m⁻² due to BC within the snow-covered areas of Himalaya and the TP, whereas Flanner et al. (2007) and Qian et al. (2011) estimated peak values of BC effects exceeding 20 W m⁻² for some parts of the TP. Menon et al. (2010) and Ménégoz et al. (2014) proposed that during the last decade BC in snow caused a significant part of the decrease of the snow cover extent or duration observed on the TP during the last decade. Ji et al. (2016b) found a positive surface RF was induced by dust, which caused a decrease of 5–25 mm w.e. over the western TP,
- 25 Himalayas, and Pamir Mountains from December to May. Large-area observations of BC data in snow cover, which are still seldom in the TP, can be useful for model evaluation and calibration in the future. To further understand the role of BC and dust in TP snow cover, more measurements and an extended LAP dataset of snow cover properties is needed to study their effects on cryospheric change in future of this region. This study is an attempt to fill this gap. Snow cover samples were collected across the TP in winter seasons (Fig. 1, Table S1), which will provide the first
- 30 large-area survey of BC and dust in seasonal snow cover over the TP, investigate albedo reduction and RF, and estimate changes of snow cover duration days caused by LAPs. Footprint analysis coupled with BC fire emission inventory will approximately constrain BC natural/anthropogenic contributions. The results of this study will increase understanding of BC and dust in snow cover across the TP and enable improved climate modeling, and this is also helpful for mitigation actions around the Tibetan cryospheric region.

3

2 Methodology

2.1 Study area

The TP covers an area of more than 2.5 million km^2 with an average elevation greater than 4000 m a.s.l. (Yao et al., 2012a). The TP exerts profound influences on atmospheric circulation and climate through mechanical and thermal effects because

- 5 of its large area, unique topography, and geographical location in the earth system (Yanai and Wu, 2006). Extended glacial coverage (over 100,000 km²) and snow cover have the potential to modify regional hydrology and trigger natural hazards that can impact local populations, agriculture, and water resources in and around the region (e.g., Immerzeel et al., 2010; Xu et al., 2009; Yao et al., 2012a).
- Moderate Resolution Imaging Spectroradiometer (MODIS) data from 2000–2006 showed that the spatial distribution of snow cover is quite variable over the TP because of the complex terrain (Pu et al., 2007). Maximum snow accumulation and melting times vary over the year but are generally later as the elevation increases. Additionally, large interannual variabilities occur in the late autumn and winter months. Heating by light-absorbing aerosols produces an atmospheric dynamic feedback, which increases moisture, cloud cover, and deep convection over northern India, and enhances the rate of snow melt in the Himalayas and the TP (Lau et al., 2010; Ramanathan and Carmichael, 2008). The accelerated melting of snow is confined
- 15 mostly to the western TP, beginning slowly in early April and then progressing more rapidly from early to mid-May; the snow cover remains reduced from mid-May through early June (Lau et al., 2010). The decadal change in the spring snow depth over the TP can impact on the Asian summer monsoon and a close relationship exists between the interdecadal increase of snow depth over the TP during March–April, increased summer rainfall over the Yangtze River valley, and a dryer summer along the southeast coast of China and the Indochina peninsula (Zhang et al., 2004). Based on observations
- from 103 meteorological stations across the TP, the duration of snow cover exhibited a significant decreasing trend (1.2 days per decade), which was jointly controlled by a later snow starting time (1.6 ± 0.8 days per decade) and an earlier snow ending time (-1.9 ± 0.8 days per decade) consistent with a response to climate change (Xu et al., 2017).

2.2 Sample collection

For this study, snow cover samples were collected at 37 sites (Fig. 1a) and one glacial river basin (Laohugou region, 25 including 10 sites as in Fig. 1b) across the TP in December 2014 and November 2015 (Table S1). As shown in Figure 1a, these field campaigns can be grouped into three regions, as was done in previous studies (Yao et al., 2013). Region I (Southern TP) includes 27 sites and is mainly affected by the Indian monsoon. Region II (Central TP) contains 10 sites that are affected by both the westerlies and the Indian monsoon. Intensified sampling (10 sites) has been carried out in the LHG region within Region III, which is located in the northeastern part of the TP. Seasonal snow samples (34) were also collected

from the 10 sites in the LHG region of Qilian Mountain (Fig. 1b) from December 2015 to June 2016.
 To minimize the effects of local sources, we selected sampling sites more than 1 km away from roads and usually 30 km from villages and cities. Generally, snow samples were collected from the top 5–10 cm with cleaned stainless-steel utensils,

4

and placed in Whirl-Pak[®] bags (Fig. S1). These snow samples were kept frozen until they underwent filtration. During sampling, snow depth, grain size, and density were also observed. For the snow grain size observation, we sprayed the snow grains on MIG paper and took a photo using a portable digital microscope (Anyty 3R-MSV500) to calculate the major and minor axis (Fig. S2). Based on field measurements from previous studies (Mugnai and Wiscombe, 1980; Grenfell and

5 Warren, 1999), two assumptions were proposed: 1) The snow grain particle is an inequilateral spheroid, and 2) The major axis (a), minor axis (b), and height (c) of the inequilateral spheroid are denoted by a, b, and c, respectively, in such a way that the relationship a=2b exists (Hao, 2009). According to Kokhanovsky and Zege (2004), the effective snow grain radius (R_{ef}) can be calculated as equal to the radius of the volume-to-surface equivalent sphere by the following equation (1):

$$R_{ef} = \frac{3V}{4\bar{A}} \tag{1}$$

10 where $\overline{V} = \frac{4}{3}\pi r^3$ and $\overline{A} = \pi r^2$, are the average volume and the average cross-section (geometric shadow) area of the snow grains, respectively. In addition, *r* is the radius of geometric optics, $r = \frac{a+b}{4}$. Thus, $R_{ef} \approx 0.35a$ (2)

2.3 LAP measurements

In the laboratory, the snow samples were melted rapidly in a hot-water bath to minimize the melt time. Immediately after melting, the snow samples were filtered through pre-baked quartz filters (WhatmanTM) using an electric pump to create a partial vacuum. Then, sample meltwater was saved in polyethylene bottles, and refrozen for chemical analysis.

Because the mineral dust mass is much larger than the BC and OC mass in the snow samples compared to aerosol samples (Wang et al., 2012), dust concentrations were calculated by taking the difference in the weight of the quartz filter before and after filtration. Quartz filters were then used to analyze BC and OC with a thermal-optical carbon analyzer (DRI 2001A

- 20 model, Desert Research Insitute, NV, USA) following a methodology (IMPROVE_A protocol) used in previous studies (Cao et al., 2003; Chow et al., 2004; Xu et al., 2009; Yang et al., 2015). Its function relies on the fact that OC components can be volatilized from the sample and deposited in a non-oxidizing helium atmosphere, in which BC can be combusted with an oxidizer. The sample punch is placed in a quartz holder oriented in the direction of the carrier gas flow, and heated gradually to a special temperature (typically 550 or 850 °C) in a pure helium atmosphere, which results in volatilization of the OC in
- 25 the sample. Then, the temperature rises and the punch is reheated stepwise to near 900 °C in an oxygen-containing atmosphere (usually 2 % oxygen and 98 % helium) to burn out all carbon remaining (BC). The carbon released from each temperature plateau is converted to methane and measured by a flame ionization detector. Due to the influence of dust on the quantitative analysis of BC and OC in snow when using a thermal optical method (Wang et al., 2012), we modified the method so that in a 100 % helium atmosphere, only a temperature plateau (550 °C) was arranged to reduce the time that BC
- 30 was exposed to a catalyzing atmosphere (Yang et al., 2015). Under ideal conditions, the minimum detection limit of total carbon is 0.93 μ g C cm⁻², with a range of 0.2–750 μ g C cm⁻². During the experiments, the filter blank (detected by blank filters) was 1.23±0.38 μ g C cm⁻².

To identify uncertainty stemming from instrumental instability and the uneven distribution of carbon particles in the filters, duplicates of ~30 % of the samples were analyzed separately, with consistent results (BC r^2 =0.95, OC r^2 =0.87, dust r^2 =0.90). Because we analyzed the filters through which the melted snow samples passed, the OC measured in our study was water-insoluble OC.

5 2.4 Footprint analysis

Calculation of air parcel trajectories is a widely used approach in different areas of atmospheric research. Conceptually, footprint analysis was considered as changes in concentration at the receptor site that can be attributed to different upwind source areas along the backward trajectories (Skiles et al., 2015). To determine the potential origins of the LAPs deposited on the snow cover of the TP, back trajectory analyses were performed using the European Centre for Medium-Range

- 10 Weather Forecasts (ECMWF) analysis fields with the Lagrangian analysis tool LAGRANTO (Sprenger and Wernli, 2015), launched every six hours for six selected sampling sites as receptors (including three sites of MYL, NMC, and SETP in region I, two sites of TGL and NETP in region II, and LHG in region III) during the winter season. The ECMWF fields (horizontal and vertical wind components) were retrieved on 137 model levels and then interpolated onto a 0.25°×0.25° latitude-longitude grid. Trajectory starting positions can be defined easily and flexibly based on different geometrical and
- 15 meteorological conditions; after the computation of the trajectories, a versatile selection is offered based on single or combined criteria (Sprenger and Wernli, 2015). First, starting positions are initialized with a suitable domain over the TP at 12:00 UTC on November 1, 2015 (or 2014). For this case, a domain from 0 to 75 and 60 °E to 120 °E was chosen. We choose starting positions in this domain that are horizontally equidistant with 80 km horizontal spacing and extend vertically from 1030 to 790 hPa with 30 hPa vertical spacing. Then, the trajectories are calculated from all starting positions 96 h
- 20 backward in time. Finally, biomass burning emission data is traced along the calculated trajectories to estimate whether an air parcel at the receptor site is influenced by BC fire emissions or not. In this study, the Fire INventory from NCAR (FINN) v1.5 global fire emissions flux in 2012–2014, speciated with the GEOS-chem mechanism, was used to estimate contributions of BC fire emission at the six selected receptor sites. FINN emission estimates are based on the framework described by Wiedinmyer et al. (2011). FINN used the satellite observations of active fires and land cover, together with
- 25 emission factors and estimated fuel loadings to provide daily, highly resolved (1 km) open biomass burning emissions estimates for use in regional and global chemical transport models. Then, the same calculation was performed for the Eclipse V5 inventory for the anthropogenic contributions of BC. The Eclipse V5 inventory was widely used in the simulations. The historical data for the period 1990–2010 were revised compared to preceding sets using the latest IAE (the International Energy Agency) and FAO (the Food and Agriculture Organization) statistics extending to 2010, as well as recent country
- 30 reporting where available. Note that this analysis was not exactly quantitative and did not take into account wet and dry deposition. We also assumed that the anthropogenic emissions have not changed significantly from 2010 compared to the period from 2012–2014. Thus, the comparison of natural and anthropogenic BC contributions was reasonable. The relative differences between the BC contributions can provide information on regional differences in this study.

6

2.5 Reflectance measurements and albedo simulations

For the selected sites, a general-purpose spectroradiometer (Analytical Spectral Devices (ASD), FieldSpec 4, Inc.), was used to measure the reflectance of snow cover. The ASD FieldSpec 4 instrument is a general-purpose spectroradiometer that is useful for applications requiring the measurement of reflectance, transmittance, radiance, or irradiance. The instrument is

- 5 specifically designed for field environment remote sensing to acquire visible and near-infrared and shortwave infrared spectra. It has 3 nm spectral resolution on the visible/near infrared detector (350–1050 nm, silicon photodiode array), and 10–12 nm resolution on the shortwave infrared detectors (900–2 500 nm, InGaAs). In the field, reflectance was measured at two sites (24K and MD in Table S1) with FieldSpec 4 under clear-sky conditions. These measurements of reflectance were calculated using the standard solar irradiance to get the albedos, which were then used for comparison with simulated
- 10 albedos.

25

Albedo reduction caused by LAPs is calculated by the SNow ICe and Aerosol Radiation (SNICAR) model (Flanner et al., 2007), which utilizes the two-stream radiative transfer solution of Toon et al. (1989) and has been widely used in Arctic snow (e.g., Flanner et al., 2007, 2009; Hadley and Kirchstetter, 2012). This simulator provides hemispheric reflectance of snow for unique combinations of impurity content (black carbon, dust, and volcanic ash), snow grain size, and incident solar

- 15 flux characteristics. The input fields include incident radiation, solar zenith angle, surface spectral distribution, effective snow grain radius, snow cover thickness and density, albedo of the underlying ground, and concentrations of impurities. Based on our observations, the effective snow grain radius ranged from approximately 100 to 1500 μ m for different snow samples based on calculation of Eq. (2), and snow density ranged from 150 to 400 kg m⁻³. These two factors varied in low, medium, and high scenarios in the model runs (Table 1).
- 20 In terms of the albedo calculation, RF due to BC and dust can be obtained by using Eq. (3) (Kaspari et al., 2014; Yang et al., 2015):

 $RF = \sum_{0.325 \ \mu m}^{2.505 \ \mu m} E(\lambda, \theta) \left(\alpha_{(r,\lambda)} - \alpha_{(r,\lambda,imp)} \right) \Delta \lambda$ (3)

where α is the modeled snow spectral albedo with or without the impurities (imp) of BC and/or dust; E is the spectral irradiance; r is the snow optical grain size; λ is wavelength (μ m); and θ is the solar zenith angle for irradiance.

- In general, the snow cover is vertically inhomogeneous, and sometimes it is optically thin (Voisin et al., 2012). The stratified structure of snow cover will lead to the discrepancy between computed spectral albedo from semi-infinite snow cover and measured albedo (Grenfell, 1994; Aoki et al., 2000; Zhou et al., 2003; Kuipers Munneke et al., 2008). In this study, we usually collected snow samples from the top 5–10 cm. Because the SNICAR model is a single-layer implementation model
- 30 (Flanner et al., 2007, 2009), the input parameter of "snowpack thickness" was the observed snow cover depth.

7

2.6 Estimates of changes in snow cover duration

To estimate snow melt due to BC and dust, a method was constructed in which the absorptivity of the snow was multiplied by the daily average incoming shortwave radiation from the automatic weather stations (AWS) set up at the meteorological stations near the sampling sites (Fig. S3) (Schmale et al., 2017). Factors derived from meteorological stations in the adjacent

5 areas included daily temperature and daily solar shortwave insolation. Average albedo reductions for snow cover by BC and dust were derived from the SNICAR model. To calculate the amount of snow melted based on the enthalpy of fusion of water (334 J g⁻¹), the snow cover was assumed to be at 0 °C and then snowmelt was calculated by using Eq. (4) (Schmale et al., 2017):

 $Melt_{snow} = N_{Tht0} \times \Delta\alpha \times SW$

10 (4)

where N_{Tht0} is the number of days with temperature greater than 0 °C; $\Delta \alpha$ is the albedo reduction caused by BC and dust, for clean snow mainly by different snow grain sizes due to snow aging; and *SW* is the shortwave radiation. From Eq. (4), we can estimate changes of snow cover duration (ΔD) based on the observed snow depth. We expressed the results for each case as low, medium, and high scenarios, based on albedo variations.

15 3 Results and discussion

3.1 Distributions of LAPs

Across the TP, most of the samples we took were old wind-packed snow. Generally, the ground layer of snow was usually depth hoar, similar to that in Arctic snow (Doherty et al., 2010) and central North American snow (Doherty et al., 2014, 2016). For many of these sampling sites, the snow cover was intermittent (Fig. S1). During the winter season, surface soil

- 20 can be carried aloft by strong winds and deposited on snow cover, which affects the concentrations of LAPs (Wang et al., 2013). Our results indicated that the spatial distributions of BC, OC, and dust in snow over the TP generally ranged from 202–17,468 ng g⁻¹, 491–13,880 ng g⁻¹, and 22–846 µg g⁻¹, respectively (Fig. 2). Variations in the concentrations of LAPs were higher in the central to northern TP (Region II and III) than in the southern TP (Region I) (Table 2). The highest concentrations may be related to the lower snow depth and dirtier snow that resulted from post-deposition processes (e.g.,
- 25 snow at some sites was melting). In Region I, the snow was cleaner and had lower concentrations of LAPs comparable to concentrations in the old snow of glaciers (Kaspari et al., 2014). The previous studies also examined concentrations of LAPs in snow from central North America (Doherty et al., 2014, 2016;

Skiles and Painter, 2016; Zatko et al., 2016), northern China (Huang et al., 2011; Wang et al., 2013; Ye et al., 2012), Japan (Kuchiki et al., 2015), Europe (Chýlek et al., 1987), the Arctic (Doherty et al., 2010; Forsström et al., 2013), Greenland

30 (Aoki et al., 2014; Zatko et al., 2013), and Antarctic (Warren and Clark, 1990; Zatko et al., 2013). Concentrations of BC and other water-insoluble LAPs from the TP were most similar to those in the snow from northern China (Wang et al., 2013), but higher than those from the Himalayas (Lim et al., 2014) (Table 2). In comparison (Table 2), the LAPs in snow cover of the

TP showed larger values which can be attributed to more deposition from nearby regions around the TP (e.g., South Asia, East Asia, and/or western China) (Lu et al., 2012; Ramanathan and Carmichael, 2008), or to impacts by the soil near the sampling sites.

- Ratios of OC to BC (OC/BC) were used to represent the possible impact of biomass burning in previous studies (Watson et al., 2001; Bond et al., 2013; Cong et al., 2015a, 2016b). Usually, the aerosols emitted from biomass burning have higher OC/EC ratios. For example, Watson et al. (2001) reported an OC/EC ratio of 14.5 for forest fires; whereas for fossil fuel, the OC/EC ratio was approximately 1. The OC/BC ratio in our sampled surface snow ranged from 0.64 to 3.31 over the TP, generally decreasing from south to north (Fig. 2d). The average ratios for Regions I, II, and III were 1.82, 1.31, and 1.14, respectively (Table 2), indicating a decreasing impact of biomass-sourced aerosol deposition in snow over the TP. The little
- 10 higher OC/BC in region I may be due to its proximity to South Asian combustion sources dominated by biomass burning (Cong et al., 2015a, 2015b; Li et al., 2016). For example, in the LHG region, lower LAP concentrations in fresh clean snow were observed at LHG3 and LHG6 (Fig. 1b), whereas near the glacier at LHG1 and LHG2, the collected wind packed, aged snow samples were quite dirty, and had higher LAP concentrations (Fig. 3a, b, and c). Meanwhile, OC/BC ratios were lower at sites near the glacier (Fig. 3d), implying predominantly non-biomass sourced combustion. Higher ratios of OC/BC were
- 15 found at the sites near human settlements (LHG7–10) (Fig. 3d), where there was extensive biofuel combustion (e.g., yak dung or straw burning). Because of the variations in sampling periods, snow types and depths, sources of dust and carbonaceous aerosol deposition, and/or strong winds, these results were expected to vary with large uncertainties. In general, however, anthropogenic activities play an important role in the ratios of OC/BC in snow cover. Figure 4 shows the footprint analyses arrived at for selected sites across the TP in winter. The northern TP sites (LHG and
- 20 NETP) are influenced mainly by air masses from Central Asia and partly by air masses from Euro-Asia, which are similar to the sources of dust reaching the TP (e.g., Zhang et al., 2016). These results are also consistent with those of He et al. (2014), Kopacz et al. (2011), and Lu et al. (2012), who modeled contributions of BC emissions in the same region. In the central (TGL and NMC) and southern region (MYL and SETP) of the TP, the air masses originate mainly from Central and South Asia. Anthropogenic BC emissions dominate the BC snow albedo as shown in previous studies (Li et al., 2016; Lu et al.,
- 25 2012). Contributions of BC from open fire burning (a kind of biomass burning) decrease from the southern to the northern TP (Fig. 5). In the Himalayan region, BC from biomass burning sources accounts for half of the BC deposition on snow cover, reflecting the proximity to large sources in South Asia. In the central TP, BC from open fire burning accounts for approximately 30 % of the total, less than from biomass burning evidence from aerosols and glacial snow (Li et al., 2016), maybe indicating a lack of biofuel contributions from our calculation. Whereas in the northern TP, anthropogenic BC
- 30 emissions (more than 70 %) dominate the BC albedo reduction. Li et al. (2016), using the $\Delta^{14}C/\delta^{13}C$ compositions of BC isolated from aerosols and snowpit samples in the TP, also found equal contributions from biomass and fossil fuel combustion in the Himalayan region (near the southern TP), whereas BC in the remote northern TP is predominantly derived from fossil fuel combustion (66±16 %). These differences can also be seen from the BC emissions that arrived at the selected sites based on BC inventory data for the year of 2013 (PKU-BC-2013, Wang et al., 2014) in Figure S3. In the southern TP

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(MYL), the amount of BC deposition is larger than in the northern TP (NETP) due to the nearby BC sources in south Asia, known to be a regional hotspot of BC-induced atmospheric solar heating (Ramanathan and Carmichael, 2008). Whereas in the central TP (NMC), the amount of BC deposition was higher than in the NETP. This may be because pollutants from the southern side of the Himalayas can traverse the high mountain range not only through the major north-south river valleys but

5 also by being lifted and advected over the Himalayas to reach to the inland TP (Cong et al., 2015b; Lüthi et al., 2015).

3.2 Impacts of LAPs on albedo reduction and radiative forcing

To simulate albedo change and surface RF from BC and dust in snow cover, the multi-layer SNICAR model was used (Flanner et al., 2007). Effective snow grain size and snow density was considered at the low, medium, and high scenarios (Table 1) and the SNICAR model was run for clear-sky conditions on the specific sampling days (Table S2).

- 10 The SNICAR model simulated albedo (caused by BC and dust with snow grain effective radius=150 μm) matches observed reflectance in both the visible and absorptive near-infrared (Fig. 6). The BC strongly reduces visible reflectance (in particular, from 350–800 nm) but has negligible influence at wavelengths beyond 1 μm, since the impact of snow grain size is large in this wavelength range (Flanner et al., 2007; Gardner and Sharp, 2010). The presence of dust also decreases the albedo in the visible wavelength, which is in agreement with a study of the impact of dust on snow radiative properties
- 15 conducted in the European Alps (Di Mauro et al., 2015). The difference between measured and simulated reflectance by dust may be caused by the upper boundary of particle size in the SNICAR model (Flanner et al., 2007). The broadband albedo is comparable to albedo simulated for the wavelengths from 350–2500 nm, with albedo differences less than 0.0058 (~2.3 %) (Table 3). For the same wavelengths (350–2500 nm), albedo between measurements and simulations differs less than 0.0155 (~4 %) (Table 3).
- For different scenarios defined by the effective snow grain size and snow density in Table 1, clean snow albedos resulting from snow aging range from 0.5992 to 0.7751 (Table 4). If BC or dust is the only impurity in the snow cover, contributions to albedo reduction can reach approximately 37 % or 15 %, respectively, with associated larger RF due to BC. On average, BC and dust contribute to albedo reduction at approximately 0.18±0.01 and 0.06±0.004, respectively (Fig. 7). The total albedo reduction contributed by BC and dust is approximately 38 %, with the associated RF of 18–32 W m⁻² (Fig. 7). This
- 25 highlights the importance of LAPs in snow when testing regional- to global-scale models. Large uncertainties exist in the estimation of BC snow albedo forcing over the TP. Simulation results using the Community Atmosphere Model version 3.1 showed that the aerosol-induced snow albedo perturbations generate surface radiative flux changes of 5–25 W m⁻² in spring (Qian et al., 2011). Using the SNICAR model coupled a general circulation model, Flanner et al. (2007) showed that, during some spring months, snow-only forcing exceeded 10 and 20 W m⁻² over parts of the TP. RF
- 30 in the snow-covered regions due to BC by the GEOS-Chem model induced the snow-albedo effect to vary from $5-15 \text{ W m}^{-2}$ over the TP, an order of magnitude larger than RF as a result of the direct effect (Kopacz et al., 2011). Recently, simulation results using a global chemical transport model in conjunction with a stochastic snow model and a radiative transfer model showed an annual mean BC snow albedo forcing to be 2.9 W m⁻² averaged over snow-covered plateau regions, which is a

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factor of three larger than the value over global land snow cover (He et al., 2014). Surface RF (simulated by a regional climate model coupled with an aerosol–snow/ice feedback module) induced by dust-in-snow showed positive values in the range of 0-6 W m⁻² over the interior of the TP, the Himalayas, Pamir, and Tienshan Mountains (Ji et al., 2016b). BC-in-snow/ice can cause positive surface RF (3.0–4.5 W m⁻²) over the western TP in the monsoon season, with the maximum RF

- 5 (5–6 W m⁻²) simulated in the Himalayas and southeastern TP in the non-monsoon season (Ji et al., 2016a). Limited measurements of LAPs in snow cover over the TP have hindered a more robust estimate of the forcing by BC and dust-in snow in this region (Bond et al., 2013; Qian et al., 2015). The mean forcing of BC and dust in snow in our study is slightly higher than previous results from snowpits (e.g., Ming et al., 2009) and comparable to some parts of the TP in spring (20 W⁻²) (Flanner et al., 2007), which indicates that more frequent impurity measurements in terms of different years and locations
- 10 over the TP are necessary for model validation. Light-absorbing OC in atmospheric aerosols has various origins, e.g., soil humics, humic-like substances (HULIS), tarry materials from combustion, bioaerosols, etc. (Andreae and Gelenscér, 2006). The chemical and optical properties of OC may differ due to the nature of the OC source or atmospheric processing (Chen and Bond, 2010). In contrast, and in comparison to black carbon and mineral dust, the optical properties of OC in snow/ice have hardly been examined. Due to the lack of
- 15 reliable OC optical properties that span the dimensions of snow grain size and OC particle size, the SNICAR model currently does not support the calculation of OC-in-snow forcing in the same way as BC. Thus, in this study, we didn't state the effect of OC on albedo reduction and RF.

3.3 Changes in snow cover duration days

Changes of snow cover duration are calculated using a model documented by Schmale et al. (2017) (section 2.6). In Eq. (4),
monthly shortwave radiation (*SW*) input data were obtained from the Automatic Weather Station (AWS) near the snow sampling sites across the TP (Fig. S3). Table S3 shows monthly shortwave radiation data from AWS during the snow melting season (March-May) when the temperature began to increase. On average, shortwave radiation in March, April, May, and June is approximately 238, 269, 292, and 271 W m⁻², respectively. Short-wave radiation in March showed the

- minimum value in Tanggula of the central TP (210 W m⁻², lower than the average), whereas in May it showed the maximum value in Namco (314 W m⁻², higher the average). Based on these data during the melt season, three scenarios (220, 270, and 310 W m⁻²)were defined as the minimum, median, and maximum scenarios of input shortwave radiation to estimate its impact on changes of snow cover duration. The average Snow depth water equivalent (SD, mm) was assumed from previous studies in this region (Che et al., 2012; Xu et al., 2017; Zhong et al., 2016). In general, the number of reduction days in snow cover duration is larger for low scenarios than for medium and high scenarios (Fig. 8 and Table 5), particularly when *SW*
- 30 input data is lower (220 W m⁻²). If SD was 40 mm, the average snow cover duration days would be reduced by 1.26±0.05 days (*SW*=310 W m⁻²) to 1.77±0.07 days (*SW*=220 W m⁻²) by BC and dust (Fig. 8a, c, and e). If SD was 100 mm, the average snow cover duration days would be shortened by 3.14±0.13 days to 4.43±0.18 days from the effect of BC and dust, with a maximum of approximately 9.4 days (Fig. 8b, d, and f), indicating advanced melt during ablation season. The result

indicated that estimation of changes of snow cover duration days caused by the same level of LAPs was also affected by different SD. For different scenarios, dust contributes only approximately 1 day to the reduction of snow cover duration days (Fig. 8 and Table 5). These results imply that the reduction in snow cover duration related to LAPs is due mainly to BC. Note that no significant differences in reductions of snow cover duration are found across the TP (Fig. S4).

- 5 Against the background of climate change, changes in duration of snow cover have important consequences for surface energy and water budgets over a range of scales, as well as for cryospheric ecosystems (e.g., Euskirchen et al., 2007; Yuan et al., 2016; Zeeman et al., 2017). Based on measurements and modeling, several studies have documented recent trends toward accelerated snowmelt and changes in snow cover duration caused by BC and dust across the Himalayan-TP in response to enhanced albedo reduction (Ménégoz et al., 2014; Menon et al., 2010; Xu et al., 2009). Ji et al. (2016a, 2016b)
- 10 noted that BC and dust in snow/ice decreases surface albedo and causes the snow water equivalent to decrease by 5–25 mm over the TP. Ménégoz et al. (2014) has estimated that both dry and wet BC depositions affect the snow cover, reducing its annual duration by 1 to 8 days, which may potentially influence sustaining seasonal water availability (Immerzeel et al., 2010; Tahir et al., 2015).

3.4 Limitations and implications for snow cover and glaciers on the TP

- 15 We calculated the effect of LAPs on snow cover duration with a range of simplistic assumptions based on simulations of albedo reduction and RF. We did not consider meteorological conditions, including clouds, precipitation, etc. Snow metamorphism was also not considered, and in the simulation, snow grains were assumed to be spherical, as previous studies (Flanner et al., 2007; Ginot et al., 2013; Schmale et al., 2017). We also did not consider the BC morphology and its mixing state, and light-absorbing properties of OC and dust, which may also significantly affect the light absorption in the
- 20 simulation (Andreae and Gelencsér, 2006; Kaspari et al., 2014; Di Mauro et al., 2015; Painter et al., 2010; Schmale et al., 2017). BC in the atmosphere tended to mix with OC and inorganic salts during aging enhancing its absorption (Gustafsson and Ramanathan, 2016). He et al. (2014) noted that BC-snow internal mixing increases the albedo forcing by 40–60 % compared with external mixing, and coated BC increases the forcing by 30–50 % compared with uncoated BC aggregates, whereas Koch snowflakes reduce the forcing by 20–40 % relative to spherical snow grains. A lack of OC simulaton due to
- 25 the limitation of the SNICAR model also affected the results in this study. Due to its light-absorbing properties (Andreae and Gelencsér, 2006; Bahaur et al., 2012; Yan et al., 2016), OC in snow cover can also absorb solar radiation and reduce the surface albedo. Previous studies indicated that OC was responsible for more than 10–40 % of the light absorption (Dang and Hegg, 2014; Doherty et al., 2010; Lin et al., 2014; Wang et al., 2015). Not considering OC in snow will underestimate the LAP impact on albedo reduction and RF, and this should not be ignored in the future.
- 30 We also have to pay attention to the fact that dust optical properties depend strongly on source material and their properties are designed to represent "global-mean" characteristics as closely as possible (Flanner et al., 2007). The light absorption by mineral dust in snow is thought to be due to iron oxides (Wang et al., 2013), which are efficient light scattering and absorption materials that can enhance absorption at UV and visible wavelengths (Di Mauro et al., 2015; Moosmüller et al.,

2012; Zhang et al., 2015). Goethite and hematite are the most abundant forms of iron oxide in dust and the major light absorbers in the shortwave spectrum in snow (Wu et al., 2016). The SNICAR model applies the Maxwell-Garnett approximation for combining indices of refractions, assuming a mixture of quartz, limestone, montmorillonite, illite, and hematite. We should note that some dust particles (e.g., those containing a large proportion of strongly absorbing hematite)

- 5 can have a larger impact on snow albedo than the dust applied in this work (Aoki et al., 2006; Painter et al., 2007). It is further important to note that a large fraction of such uncertainty can also result from uncertainty in the simulated snow-cover fraction/snow depth across the plateau. The TP covers a large area (more than 2.5 million km²) with an average elevation exceeding 4000 m a.s.l. (Yao et al., 2012a). Considerable heterogeneity in the topography and climate has led to complex spatial and temporal snow cover patterns (Xu et al., 2017). Most studies on the snow cover distribution were based
- 10 on satellite-based observations (Che et al., 2008, 2012). Surface observations of snow depth showed an increase over the TP from 1957–1998 (Ma and Qin, 2012). However, snow cover depth and the number of snow covered days during the current decade under intense climate warming showed a negative trend mainly occurring in the southeast TP in winter. Whereas in spring, snow cover depth showed a positive trend in the eastern TP (Xu et al., 2017). These differences can also affect the estimation in this study. Nevertheless, the method provides a theoretical approach for evaluating how the presence of LAPs affects the lower parts of the glacier subjected to summer melt.
- Snow cover durations were shortened during the melt season as estimated in this study, and the glaciers over western China have been undergoing extensive recession, with an estimated loss of about \sim 450 km³ w.e. in volume and 10 % reduction in area during the last 40 years (Yao et al., 2012b). Recently, the annual mass budget of glaciers over the TP was estimated at approximately 15–20 Gt yr⁻¹ based on GRACE and ICESat (Gardner et al., 2013; Neckel et al., 2014). These melt waters
- 20 may affect water resources and social development in the surrounding areas, e.g., India and China (Immerzeel et al., 2010). The total amount of snowmelt caused by LAPs can be estimated over the whole TP using the model described in Sec. 2.6. The number of days with average temperature above 0 °C was assumed to be 70 (± 25 %) based on the observations from AWS during the melting season. Thus, based on the daily snowmelt (0.76 \pm 0.40, 0.67 \pm 0.36, and 0.54 \pm 0.28 cm w.e. d⁻¹ for SW at 220, 270, and 310 W m⁻², respectively) caused by BC and dust in this study, the estimation shows that approximately
- 4.61 \pm 2.49 Gt yr⁻¹ (range of 3.80 \pm 1.97 to 5.36 \pm 2.78 Gt yr⁻¹) of snowmelt water has been lost due to the effect of BC and dust in snow (Fig. 9). This indicates that glacial snowmelt caused by LAPs contributes ~20 % of total mass balance and that BC and dust may play an important role in recent accelerated snow and glacial melt. However, this result has large uncertainties associated with the lack of sufficient *in situ* and distributed observations of spectral albedo, concentrations of LAPs, and coincident changes in glacier mass observations, as discussed by many previous studies (e.g., Ginot et al., 2014; Ménégoz et
- 30 al., 2014; Qian et al., 2015; Schmale et al., 2017).

Our study confirms that BC and dust in snow and ice can darken the surface, and further reduce snow albedo, and increase the speed of snow cover melt (e.g., Flanner et al., 2007; Di Mauro et al., 2015; Painter et al., 2007; Qian et al., 2015; Skiles et al., 2015). Thus, the impacts of BC and other LAPs need to be properly accounted for in future regional climate projections, particularly in the high-altitude cryosphere. It is also important to note that, without large scale, *in situ*

observation data applied to calibrate BC forcing and constrain simulated values, the impact of BC and other LAPs on the albedo feedback would have been marginal. Schmale et al. (2017) noted that, due to the decreasing trend of dust emissions in Central Asia (Xi and Sokolik, 2015), dust may become less important, while BC deposition may increase as a result of projected increased emissions (Wang et al., 2014). Constraints on BC sources/deposition and climatic/hydrological effects will provide guidence for offseting mitigation entions.

5 will provide guidance for effective mitigation actions.

4 Conclusions

This study was a large area survey of snow cover over the TP across regions with different climatology and sources of LAPs. Systematically higher values of snow BC concentrations were observed in the central to northern TP (Regions II and III) than in the southern TP (Region I). This finding may be related to shallower snow depths and dirtier snow in Regions II and

- 10 III as a result of post-depositional processes (e.g., snowdrift, or melting). In the southern TP (Region I), the collected snow was cleaner with lower concentrations of LAPs comparable to concentrations in the glacial aged snow. The ratios of OC/BC in the surface snow samples ranged from 0.64 to 3.31 across the TP, generally decreasing from south to north, which may indicate that anthropogenic activities played an important role on OC/BC ratios in snow cover in the TP region. Footprint analysis also indicated that anthropogenic sourced BC accounted for approximately 70 % of total BC deposition in the
- 15 central and northern TP. Estimates of albedo reduction contributed by BC and dust were approximately 38 %, with an associated RF of $18-32 \text{ W m}^{-2}$.

The reported shortage of snow cover duration days across the TP relied on single site observations rather than a large area survey. For different scenarios with different SD and shortwave radiation input data, the advanced melt days of snow cover duration related to LAPs was mainly due to BC, which reduced from several days to more than one week. Dust contributed

20 only approximately 1 day to the reduction of snow cover duration days. No significant differences among reductions in snow cover duration were found across the whole TP. The changing snow cover duration may potentially influence seasonal water availability and sustainability.

Furthermore, the estimated effects of BC and dust on glaciers and snowmelt across the TP were based on the simulations of albedo reduction and RF. It was found that glacial snowmelt water caused by LAPs contributed ~20 % of the total mass

25 balance. These findings revealed that the effects of both BC and non-BC absorbers need to be properly accounted for in future regional climate projections, in particular on the high-altitude cryosphere.

Data availability

Black carbon and other light absorbing data are available upon request from Professor Shichang Kang and Dr. Yulan Zhang.

Author contribution

30 Y. Zhang, S. Kang, and Z. Cong designed the experiments. M. Sprenger performed the footprint simulations. Y. Zhang, T. Gao, C. Li, X. Li, X. Zhong, and M. Xu, carried out the experiments. S. Tao and W. Meng analyzed the black carbon

inventory. M. Sillanpää and X. Qin gave valuable comments. Y. Zhang prepared the manuscript with contributions from all co-authors.

Acknowledgement

We acknowledge the support provided by the National Natural Science Foundation of China (41630754, 41671067, and

5 41421061), State Key Laboratory of Cryospheric Sciences (SKLCS-ZZ-2017), Chinese Academy of Sciences (KJZD-EW-G03-04), the Foundation for Excellent Youth Scholars of the Northwest Institute of Eco-Environment and Resources, CAS, and the China Scholarship Council. We also thank Dr. M Flanner for help with simulating reflectance with the SNICAR model, and the reviewers who helped to improve this manuscript.

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Zhou, L., Dickinson, R. E., Tian, Y., Zeng, X., Dai, Y., Yang, Z.-L., Schaaf, C. B., Gao, F., Jin, Y., Strahler, A., Myneni, R. B., Yu, H., Wu, W., Shaikh, M.: Comparison of seasonal and spatial variations of albedos from Moderate-Resolution Imaging Spectradiomater (MODIS) and Common Land Model, J. Geophys. Res, 108(D15), doi:10.1029/JD003326, 2003.

4 Table 1 Snow effective grain sizes and snow density used for the albedo calculation with the SNICAR model.

Description	Low scenario	Central scenario	High scenario
Snow effective grain radius (µm)	150	500	1500
Snow density (kg m ⁻³)	150	250	400

Sites	BC (ng g^{-1})	Dust (µg g ⁻¹)	OC (ng g^{-1})	OC/BC	Method	References
Southern TP (Region I)	1423±242	119.8±18.97	2145±325	1.82±0.17	DRI	This study
Central TP (Region II)	5624±1500	295±67.50	6119±1257	1.31±0.19	DRI	This study
Northern TP (Region III)	1484±626	93.20±27.05	974±197	1.14±0.15	DRI	This study
Himalayan Khumbu	0.1-70				SP2	I
Valley	0.1-70				SP2	Jacobi et al., 2015
Himalayas	24.3±20.1	1.32±0.84	359±185.2	~10	Sunset	Lim et al., 2014
Qilian mountain	1550				ISSW	Wang et al., 2013
Border of Siberia, China	117				ISSW	Wang et al., 2013
Northeast China	1220				ISSW	Wang et al., 2013
Inner Mongolia, China	340				ISSW	Wang et al., 2013
Northwest China	10-150				ISSW	Pu et al., 2016
Sapporo, Japan	7-2800	10-1300	0.14-260		Sunset	Kuchiki et al., 2015
Central North America	5-70				ISSW	Doherty et al., 2014
Uintah basin, Utah, USA	5-100				ISSW	Zatko et al., 2016
Sierra Nevada, North	11.77				DDI	
America	11±7.7				DRI	Hadley et al., 2010
French Alps	4.3±4.5	3.06±5.25	59.6±77.6		SP2	Lim et al., 2014
Greenland Summit	3.1±1.4	0.262±0.117	142.6±82.6		SP2	Lim et al., 2014
Greenland	Sumit station: 1.4				ISSW	Zatko et al., 2013
	Scandinavia: 88					
	Svalbard: 11-14				G (E 1 . 0010
Arctic	Fram Strait: 7-42				Sunset	Forsström et al., 2013
	Barrow: 9					
	Near Dome C: 2.1				ICON	
Antarctic	Far from Dome C: 0.6				ISSW	Zatko et al., 2013

6 Table 2 Summary of BC, OC and dust concentrations in snow over the Tibetan Plateau and other regions.

8 (1) DRI-DRI 2001A model thermal-optical carbon analysis

9 (2) SP2: Single Particle Soot Photometer

(3) ISSW- ISSW spectrophotometer 10

11 (4) Sunset: Sunset Lab OC-EC Aerosol Analyzer

12

13 Table 3 Comparison between measured albedo by ASD and simulated albedo (SA) by the SNICAR model.

Selected site (24K)	SA clean snow	SA BC	SA Dust	SA BC+Dust	
SNICAR: Broadband (305-4495 nm)	0.7824	0.7027	0.7511	0.6892	
SNICAR: Wavelength (350-2500 nm)	0.7728	0.6985	0.7453	0.6864	
ASD measurement (350-2500 nm)		0.6709±0	0.0019		

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Table 4 Simulated albedo (SA) and radiative forcing (RF) by BC and dust in snow cover across the Tibetan

17 Plateau.

Different scenarios	SA clean snow	SA BC	SA Dust	SA BC+Dust	Contribution to albedo reduction	RF BC	RF Dust	RF BC+Dust
Low	0.7751±0.0193	0.6517 ± 0.0702	0.7387±0.0250	0.6413±0.0710	37 %	16.32±0.0931	4.78±0.0310	17.64±0.0952
Medium	0.6943±0.0313	0.5104 ± 0.0900	0.6372 ± 0.0382	0.4992 ± 0.0900	39 %	24.20±0.1250	7.57±0.0483	25.86±0.1233
High	0.5992±0.0435	0.3683±0.0935	0.5197±0.0490	0.3582±0.0889	38 %	30.96±0.1362	10.70±0.0653	32.36±0.1315

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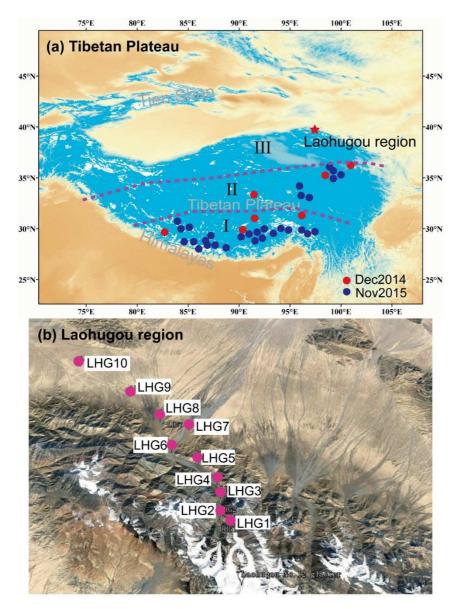
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20 Table 5 Average reductions of snow cover duration by BC and dust for different short wavelengths (220,

21 270, and 310 W m⁻², respectively) and snow cover depth water equivalent (mm).

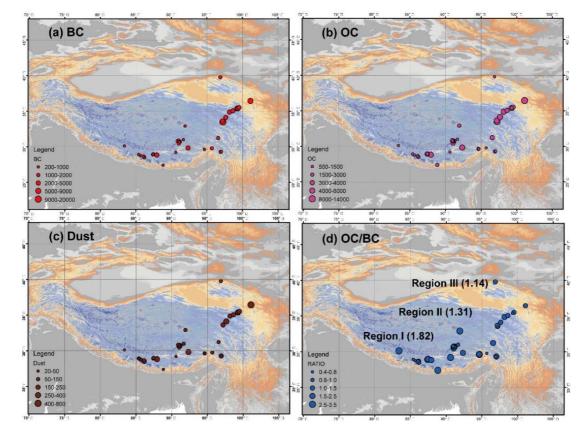
SD	SD=40 mm			SD=100 mm			
Shortwave	BC	Dust	BC+dust	BC	Dust	BC+dust	
SW=220 W m ⁻²	1.69 ± 0.07	0.74±0.04	1.77±0.07	4.24±0.18	1.84 ± 0.10	4.43±0.18	
SW=270 W m ⁻²	1.38±0.06	0.60 ± 0.03	1.44 ± 0.06	3.45±0.15	1.50 ± 0.08	3.61 ± 0.15	
SW=310 W m ⁻²	1.20 ± 0.05	0.52±0.03	1.26±0.05	3.01±0.13	1.31±0.07	3.14±0.13	

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25 Figure 1: Snow sampling sites over (a) the Tibetan Plateau and (b) the Laohugou region.

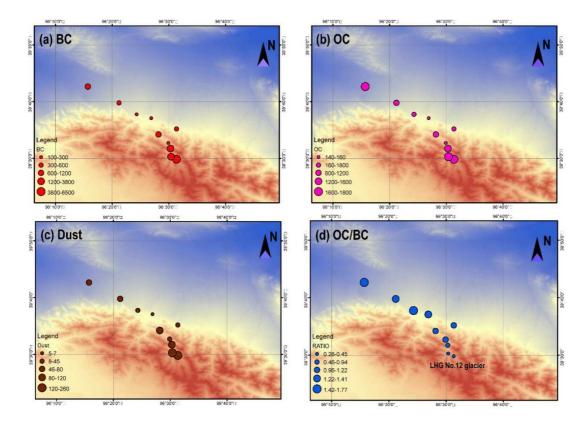


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28 Figure 2: Spatial distributions of light-absorbing impurities in snow cover for each sampling site over the

29 Tibetan Plateau. (a) BC, (b) OC, (c) Dust, and (d) ratio of OC to BC (OC/BC).



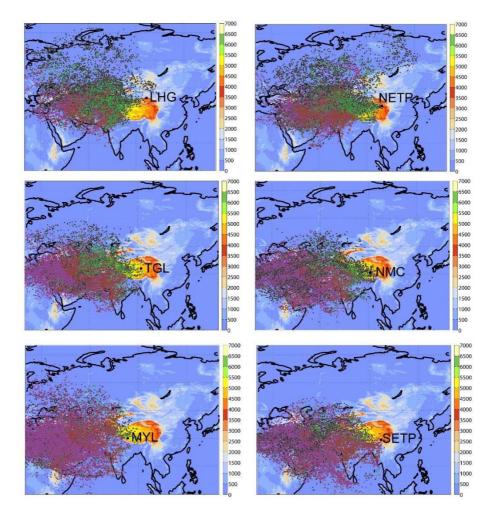


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32 Figure 3. Spatial distributions of light-absorbing impurities in snow at the Laohugou region. (a) BC, (b) OC,

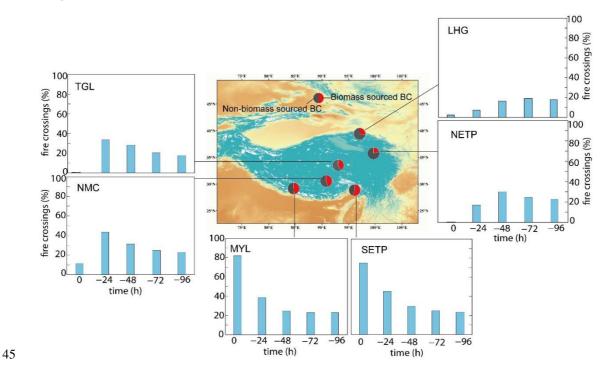
33 (c) Dust, and (d) ratio of OC to BC (OC/BC).



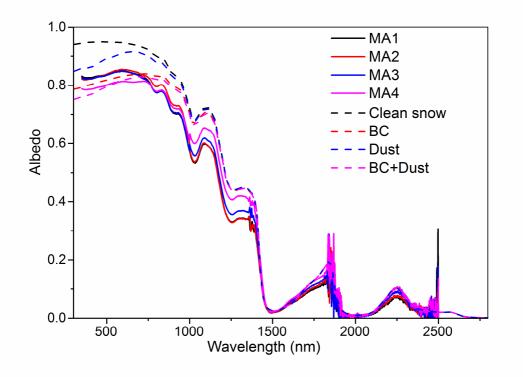


36 Figure 4: Footprint analyses for six selected sites over the Tibetan Plateau during the winter season (Nov 37 2015-Feb 2016). LHG and NETP in the northern Tibetan Plateau (Region III), TGL and NMC in the 38 central Tibetan Plateau (Region II), and MYL and SETP in the southern Tibetan Plateau (Region I). (The 39 right color bar means the height (m a.s.l.). The trajectories starting below 500 hPa were taken into account. 40 Black dots: the air parcel did not pass near a fire during the 96 h prior to arrival at the studied site; Green 41 dots: the air parcel did pass by a fire between -96 h and -48 h, but not afterwards, i.e., the 'contact' to a fire 42 lies back at least 48 h before the air parcel arrived at Renlongba glacier; Magenta dots: contact with fires 43 occurred between -48 h and 0 h before arriving at site; Red dots: contact with a fire before and after -48 h.)

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46 Figure 5: Different source contributions to BC deposition on the snow cover of the Tibetan Plateau.

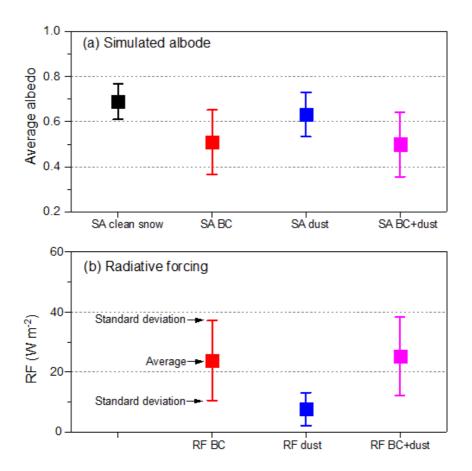


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49 Figure 6: Measured albedo by ASD (solid lines, MA1-4) and simulated effects of BC and dust on albedo

50 (dashed lines) at the selected snow site on the Tibetan Plateau.





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Figure 7: (a) Average albedo (solid rectangles) of snow cover due to aging only and impurities relative to the clean snow and (b) resulting clear sky instantaneous radiative forcing (solid rectangle means the average). (Note: SA means simulated albedo, SA BC means albedo only with BC in snow, SA dust means albedo only with dust in snow, and SA BC+dust means albedo with BC and dust in snow. RF means radiative forcing, RF BC means the RF caused by BC in snow, RF dust means the RF caused by dust in snow, and RF BC+dust means the RF caused by BC and dust in snow. The solid rectangles are the central average estimate whereas the bars show the standard deviation range of albedo and RF)

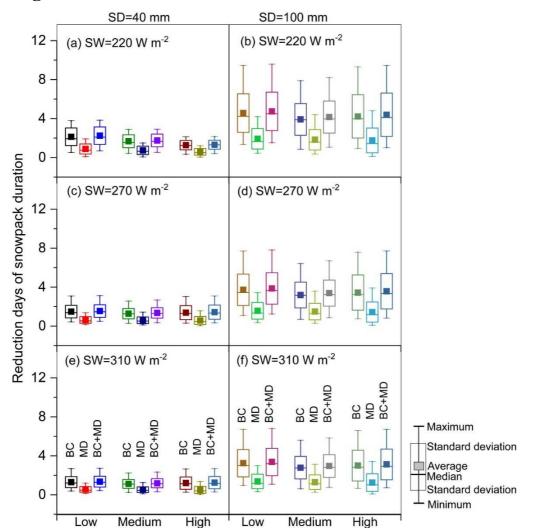
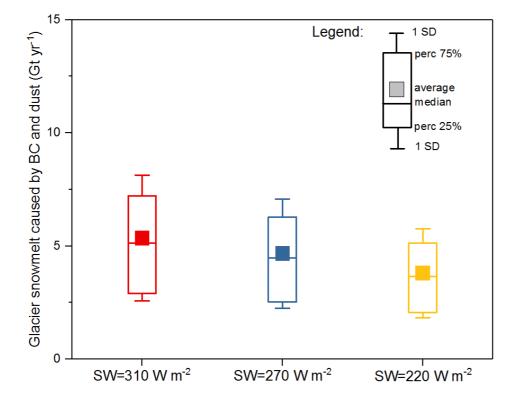


Figure 8: Reduction in days of snow cover duration by BC and dust for low, medium and high scenarios due to albedo reduction at different snow depth (SD=40 and 100 mm, respectively). a and b were simulated based on the daily shortwave at 220 W m-2. c and d were simulated based on the daily shortwave at 270 W m-2, e and f were simulated based on the daily shortwave at 310 W m-2. (Solid rectangles are average values; solid line in the box means the median; outliers of boxes are the one standard deviations of the average; and the bars are maximum and minimum values.)





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Figure 9: Estimates of total glacial snowmelt based on daily snowmelt caused by BC and dust for shortwave
radiation at 220, 270, and 310 W m⁻², respectively. The days of daily temperature above 0 ° C value is 100
(±25 %).

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