

Response to Referee #1

We are very grateful for the referee's critical comments and suggestions, which have helped us improve the paper quality substantially. We have addressed all of the comments carefully as detailed below in our point-by-point responses. Our responses start with "R:".

This paper uses snow sample observations across northern China in January 2014 and aerosol radiation models to examine the reduction of snow albedo due to black carbon, organic carbon and anthropogenic dust. The study suggests different contributions to the snow albedo reductions from these aerosols and suggests that biomass burning may be a major contributor at most snow sampling sites. It also evaluates the model simulations based on the observations. In general, this paper provides useful information, particularly the different performance in snow albedo reduction by three types of aerosols. However, there are still a few limitations. The small sample volume observed over short period at limited sites could make the results questionable. At least discussions about the uncertainties and potential issues in this study are necessary. Corresponding to the limited data, reliable or strong quantitative conclusion is hard to obtain. The authors could provide more quantitative results and discuss the reliability of the findings in the study.

R: We agree with the reviewer that the number of snow sample is small from the only 13 monitoring sites. One reason for the small number of samples in this season (2014) was due to the less snowfall across northern China. Despite with the small snow data samples, we have designed the study carefully by running two models and combing with various other data sets to generate new scientific knowledge. The importance of our study can be reflected in the following aspects:

(1) Seasonal snow amount at mid-high latitude regions has large spatial variations in any given year. One novelty of the present study is to investigate the spatial and vertical variations of BC in seasonal snow and attribute the light absorption to BC, OC and mineral dust.

(2) Several recent studies have indicated that the mixing states of BC and the irregular morphology of snow grain have large effects in snow albedo reduction (He et al., 2014; Liou et al., 2011, 2014). Based on the comments from this reviewer and from the interactive comments provided by Cenlin He, we have provided a new figure in the revised paper discussing the uncertainties in the mixing states and the irregular morphology of snow grains on snow albedo reduction estimation using SAMDS model simulations (Figure 10).

(3) The title has been changed as "Observations and model simulations of snow albedo reduction in seasonal snow due to insoluble light-absorbing particles during 2014 Chinese survey".

The manuscript is totally rewritten, and more results in discussing the SAMDS for snow albedo reduction due to ILAPs in snow, internal/external mixed BC in snow, and the snow grain shapes were given in the abstract, introduction, methods, results, and the conclusions. Details have already been illustrated in comment 1.

Are the snow samples fresh or aged snow?

R: We have already updated this information in Table 1 and Figure 1.

(2) Snow albedo reduction due to these kinds of aerosols is well known, more quantitative results are needed and more valuable, which could be summarized in the abstract. At current version, it is a little hard for me to summarize the findings I can learn from this paper.

R: We agree with the reviewer that optical properties of black carbon in snow have been investigated in earlier studies (e.g. Liou et al., 2014; Qian et al., 2014; Warren et al., 1980). However, very few studies have focused on light absorption by OC and dust in snow, which is one of the focuses of our study here. We have investigated snow albedo reduction by OC and mineral dust using the SNICAR model and a new SAMDS model. The impact of the uncertainties in the mixing states and the irregular morphology of snow grain on albedo reduction is also illustrated using SAMDS model simulations (Figure 10)..

Page 3, line 11-14, why is Ginoux et al. 2010 cited two times in one sentence (begin and end). This is repeated description and one should be deleted.

R: We have deleted the second citations of Ginoux et al., (2010), and we have also corrected the similar mistakes throughout the manuscript.

Page 4, line 4-5, you use “larger” and “more intense” in the sentence, but I did not see any comparison descriptions around this sentence. What you are comparing?

R: We have added a reference of Guan et al. (2016), which illustrated the relationship between anthropogenic dust and population over global semi-arid regions, and the source attribution of insoluble light-absorbing particles in seasonal snow across northern China. Furthermore, Wang et al. (2013a) also indicates that BC emission sources in China are strongest in far eastern China of our northeast China snow sampling than the other regions across northern China (Figure 11 in Wang et al., 2013a).

Page 4, Line 12-13, why is Light et al. 1998 cited two times in one sentence (begin and end). This is repeated description and one should be deleted.

R: Corrected.

Page 5, line 15-24, you mentioned several campaigns for snow collection. What are the differences or similarities for the findings among them? Also, it shows that there is a snow campaign over the examined region in 2010 carried out by Huang et al. 2011. Why do not you also include the observation from this campaign so that you have enough data samples and you can also compare the differences/similarities in two winters?

R: In this manuscript, we try to focus on the new findings due to ILAPs in snow during 2014 field campaigns more clearly. As a result, we deleted figure 6 and section

3.3. Figure 4 was also modified as figure 4a and 4b based on 2014 snow field campaigns, which weren't published in previous studies.

As Doherty et al., (2015) indicated that with no measure of the interannual variability of the mixing ratio of BC in snow, it is difficult to determine the representativeness of the samples collected in the Arctic survey.

R: We agree with the reviewer. For this reason, we note that further field campaigns on measuring mixing ratio of BC in snow should be performed worldwide in northern Hemisphere. However, these datasets were much useful for climate models to reduce the uncertainty of the climate effects due to BC in snow.

Page 7, line 1-3, is this data criteria enough to prevent contamination? And why do you include site 101 if it does not fit your data criteria?

R: We indicate that the datasets used in this study are criteria enough to prevent contamination. The reason is that the snow sample collection and the analysis procedure were strictly performed followed by Doherty et al. (2010) and Grenfell et al. (2011). Although site 101 is close to the village, we point out that the ILAPs in snow are more representative for the country village regions across northern China.

Page 7, line 5-10, what is the uncertainty introduced due to your visual inspection and data processing method?

R: As shown in Grenfell et al. (2011), visual comparison is best carried out under diffuse reflected illumination with the filters sitting on a white diffusing background. Uncertainties including personal bias involved on measuring BC in snow is approximately a factor of ~1.5-2. The causes of the bias in the visual estimates of the China 2010 field filters vs. those from the expedition reported by Grenfell et al. (2011) is unknown (Grenfell et al., 2011; Wang et al., 2013a).

Page 8, line 1-9, what do some variables (not all) stand for?, such as Ss?

R: The subscript of Ss means sea salt sources. We have added explanations for all the variables that were not labeled in this manuscript.

Page 8, line 17, why do not you introduce Fe when iron is first used in paper?

R: We indicated that Fe in this manuscript is the same as iron (Page 9, line 17) . Another consideration is that when discussing chemical elements, the symbol Fe instead of iron is commonly used.

Page 9, line 4-6, How do you know Microtops II is reliable, or more reliable than CE318?

R: We indicated that Microtops II and CE318 are both effective instruments on measuring aerosol optical depth (AOD) (More et al., 2013; Porter et al., 2001; Zawadzka et al., 2014). However, the major difference between Microtops II and CE318 is that the Microtops II is portable for the field experiments, but CE318 is immovable. For the snow survey, it is better to use the Microtops II instrument

instead of CE318 to measure aerosol optical depth.

Page 9, Line 22, do you mean “in 2014”?

R: We have already corrected this sentence as “Fire locations were based on data provided by the MODIS FIRMS system from October 2013 to January in 2014.”

Page 10, line 4-5 and line 19-20, why do the observations and models calculate the albedo at different height (1 m above snow vs at surface)? Also, the downwelling solar radiation in the real sky includes diffuse radiation. How does the model consider the diffuse radiation (such as the contribution from aerosols and clouds in the sky)?

R: Normally the relative position of the sighting laser spot is at a distance of 1m from the optical element for the active field of view for the instrument in strict accordance with the user manual of the SVC HR-1024 spectroradiometer (Figure 6 Setup for FOV map). The direction of the instrument was oriented to the Sun Horizon angles in order to receive more direct solar radiation. The small size of the fore optics greatly reduces errors associated with instrument self-shadowing. Even when the area viewed by the fore optic is outside the direct shadow of the instrument, the instrument still blocks some of the illumination (either diffuse skylight or light scattered off surrounding objects) that would normally be striking the surface under observation for measuring full-sky-irradiance throughout the entire 350 - 2500 nm wavelengths. This spectroradiometer is used for measuring the direct component of solar irradiance because of the minimized relative radiometric errors between total and direct irradiance measurements. For instance, Bi et al. (2013) used a set of broadband radiometers and sun/sky photometers during 2013 field campaign in the middle latitude across northern China to measure the direct and diffuse solar irradiance, and the result indicated that the diffuse solar radiation is 10% lower than the total solar irradiance. Therefore, we indicated that the spectroradiometer in the clean sky condition mainly measured the direct solar irradiance during 2014 snow campaign. The above materials have been added in section 2.4 of the revised paper to present the relative parameter of the spectroradiometer.

Page 11, line 15, the symbol should be μ , please explain its meaning.

R: We have changed the symbol as μ in Page 14, line 16, and μ refers to the escape function in radiative transfer theory (Kokhanovsk and Zege, 2004).

Page 15, line 7-10, both AOD from MODIS and ground are retrieved, please describe clearly.

R: The ground AOD is retrieved by Microtops II sun photometer. We have already indicated that Microtops II sun photometer is an effective instrument on measuring AOD. However, the weakness of this instrument is that it can only precisely measure AOD during the clean sky. So we only measured the ground AOD dataset in six sites. Then we used the MODIS AOD dataset to compare with our ground measurements to indicate the spatial variations of AOD across northern China. The active open fire retrieved from MODIS was also used to show the possible sources of the BC and OC.

Page 15, line 16, colder -> cold

R: corrected.

Page 15, line 23, This sentence is not complete. I believe what you want to say is “..., which with ...”

R: This sentence has been modified as: “In Inner Mongolia, the snow cover was thin and patchy. The average snow depth at sites 90, 91, 93, and 94 was less than 10 cm, which was significantly smaller than those (13 to 20 cm) at sites 95-97 near the northern border of China.” in page 18 lines 20-24.

Page 16, line 2-4, “Because less snow fell during the 2014 snow survey period, the surface snow grain radius varied considerably from 0.07 to 1.3 mm.”. First, I do not understand this causal relationship, please clarify. Second, what does it compare when using ‘less’, the same period of other years or other locations?

R: We have revised the sentence as: “The maximum snow depth was found to be 46 cm at site 102 inside a forest near the Changbai Mountains. Snow depth varied from 13 to 46 cm at sites 98 to 102 with an average of 27 cm. R_m of the snow samples varied considerably from 0.07 to 1.3 mm. R_m increased with the snow depth from the surface to the bottom, larger than previously recorded because of snow melting by solar radiation and the ILAPs.” in page 19 lines 5-10.

Page 16, line 19-24, why the variations (BC and snow spatial distribution) you found were much higher than the findings from other studies?

R: BC in snow in this manuscript was mostly collected in heavy industrial regions in northern China, where the mixing ratios of BC and OC were much higher than in the other regions of northern China (Flanner et al., 2007; Wang et al., 2013a; Zhao et al., 2014).

Page 17, line 1-8, I cannot catch the main points you would like to deliver here.

R: This section has been completely rewritten for clarification.

Page 17, Figure 5, the sample volume is too small. How reliable are the relationships found here?

R: We have added the datasets of BC measurements in seasonal snow during 2010 field campaign in Figure 5. The caption of Figure 5 was rewritten as “Comparisons between the calculated and optically measured C_{BC}^{est} in surface snow during 2010 and 2014 snow surveys. The datasets of measured C_{BC}^{est} in 2010 from sites 3-40 were reprinted from Wang et al. (2013a).”. We have also provided the confidence test of the fitting in Figure 5.

Page 17, line 22-24, I do not understand from Figure 6 how you got this range in OC/BC ratio.

R: See our answer to the next question.

Page 18, line 1-16, it is hard to conclude due to limited data and sites.

R: Following the reviewer's suggestions, we chose to concentrate on the ILAPs in snow and the snow albedo reduction due to internal/external ILAPs in snow, and the snow grain shapes. Therefore, we have deleted figure 6, and added a new figure in discussing the snow albedo reduction due to internal/external mixed BC in snow and difference snow grain shapes as Figure 10 in this revised manuscript.

Page 18, line 18-22. How did you get the observation regarding land-cover type when the land is covered by snow?

R: The land-cover types (Figure 7) were obtained from the Collection 5.1 MODIS global land-cover type product (MCD12C1) at a 0.05° spatial resolution. The dataset included 17 different surface vegetation types (Friedl et al., 2010; Loveland and Belward, 1997). What we wanted to demonstrate is that most of the sampling regions were correlated with human activities, while this manuscript mainly focused on the anthropogenic dust and the other ILAPs in seasonal snow.

(25)Page 19, Line 17-20, why is Wang et al. 2015 cited two times in one sentence (begin and end). This is repeated description and one should be deleted.

R: Corrected.

References:

- Bi, J. R., Huang, J. P., Fu, Q., Ge, J. M., Shi, J. S., Zhou, T., and Zhang, W.: Field measurement of clear-sky solar irradiance in Badain Jaran Desert of Northwestern China, *J. Quant. Spectrosc. Ra.*, 122, 194-207, 10.1016/j.jqsrt.2012.07.025, 2013.
- Doherty, S. J., Steele, M., Rigor, I., and Warren, S. G.: Interannual variations of light-absorbing particles in snow on Arctic sea ice, *J. Geophys. Res.-Atmos.*, 120, 11391-11400, 2015.
- Doherty, S. J., Warren, S. G., Grenfell, T. C., Clarke, A. D., and Brandt, R. E.: Light-absorbing impurities in Arctic snow, *Atmos. Chem. Phys.*, 10, 11647-11680, 2010.
- Flanner, M. G., Zender, C. S., Randerson, J. T., and Rasch, P. J.: Present-day climate forcing and response from black carbon in snow, *J. Geophys. Res.-Atmos.*, 112, D11202, 2007.
- Friedl, M. A., Sulla-Menashe, D., Tan, B., Schneider, A., Ramankutty, N., Sibley, A., and Huang, X. M.: MODIS Collection 5 global land cover: Algorithm refinements and characterization of new datasets, *Remote Sens. Environ.*, 114, 168-182, 2010.
- Ginoux, P., Garbuzov, D., and Hsu, N. C.: Identification of anthropogenic and natural dust sources using Moderate Resolution Imaging Spectroradiometer (MODIS) Deep Blue level 2 data, *J. Geophys. Res.-Atmos.*, 115, D05204, 2010.
- Grenfell, T. C., Doherty, S. J., Clarke, A. D., and Warren, S. G.: Light absorption from particulate impurities in snow and ice determined by spectrophotometric analysis of filters, *Appl. Opt.*, 50, 2037-2048, 2011.

- Guan, X. D., Huang, J. P., Zhang, Y. T., Xie, Y. K., and Liu, J. J.: The relationship between anthropogenic dust and population over global semi-arid regions, *Atmos. Chem. Phys.*, 16, 5159-5169, 2016.
- He, C. L., Li, Q. B., Liou, K. N., Takano, Y., Gu, Y., Qi, L., Mao, Y. H., and Leung, L. R.: Black carbon radiative forcing over the Tibetan Plateau, *Geophys. Res. Lett.*, 41, 7806-7813, 2014.
- Huang, J. P., Fu, Q. A., Zhang, W., Wang, X., Zhang, R. D., Ye, H., and Warren, S. G.: Dust and Black Carbon in Seasonal Snow across Northern China, *Bull. Amer. Meteor. Soc.*, 92, 175-181, 2011.
- Kokhanovsky, A. A., and Zege, E. P.: Scattering optics of snow, *Appl. Opt.*, 43, 1589-1602, 2004.
- Liou, K. N., Takano, Y., and Yang, P.: Light absorption and scattering by aggregates: Application to black carbon and snow grains, *J. Quant. Spectrosc. Ra.*, 112, 1581-1594, 2011.
- Liou, K. N., Takano, Y., He, C., Yang, P., Leung, L. R., Gu, Y., and Lee, W. L.: Stochastic parameterization for light absorption by internally mixed BC/dust in snow grains for application to climate models, *J. Geophys. Res.-Atmos.*, 119, 7616-7632, 2014.
- Loveland, T. R., and Belward, A. S.: The IGBP-DIS global 1 km land cover data set, DISCover: first results, *Int. J. Remote Sens.*, 18, 3291-3295, 1997.
- More, S., Kumar, P. P., Gupta, P., Devara, P. C. S., and Aher, G. R.: Comparison of Aerosol Products Retrieved from AERONET, MICROTOPS and MODIS over a Tropical Urban City, Pune, India, *Aerosol. Air. Qual. Res.*, 13, 107-121, 2013.
- Porter, J. N., Miller, M., Pietras, C., and Motell, C.: Ship-based sun photometer measurements using Microtops sun photometers, *J. Atmos. Oceanic Technol.*, 18, 765-774, 2001.
- Qian, Y., Wang, H. L., Zhang, R. D., Flanner, M. G., and Rasch, P. J.: A sensitivity study on modeling black carbon in snow and its radiative forcing over the Arctic and Northern China, *Environ. Res. Lett.*, 9, 064001, 2014.
- Wang, X., Doherty, S. J., and Huang, J. P.: Black carbon and other light-absorbing impurities in snow across Northern China, *J. Geophys. Res.-Atmos.*, 118, 1471-1492, 2013a.
- Wang, X., Pu, W., Zhang, X. Y., Ren, Y., and Huang, J. P.: Water-soluble ions and trace elements in surface snow and their potential source regions across northeastern China, *Atmos. Environ.*, 114, 57-65, 2015.
- Warren, S. G., and Wiscombe, W. J.: A Model for the Spectral Albedo of Snow .2. Snow Containing Atmospheric Aerosols, *J. Atmos. Sci.*, 37, 2734-2745, 1980.
- Yasunari, T. J., Koster, R. D., Lau, W. K. M., and Kim, K. M.: Impact of snow darkening via dust, black carbon, and organic carbon on boreal spring climate in the Earth system, *J. Geophys. Res.-Atmos.*, 120, 5485-5503, 2015.
- Zawadzka, O., Makuch, P., Markowicz, K. M., Zielinski, T., Petelski, T., Ulevicius, V., Strzalkowska, A., Rozwadowska, A., and Gutowska, D.: Studies of aerosol optical depth with the use of Microtops II sun photometers and MODIS detectors in coastal areas of the Baltic Sea, *Acta Geophysica*, 62, 400-422, 2014.
- Zhao, C., Hu, Z., Qian, Y., Leung, L. R., Huang, J., Huang, M., Jin, J., Flanner, M. G., Zhang, R., Wang, H., Yan, H., Lu, Z., and Streets, D. G.: Simulating black carbon and dust and their radiative forcing in seasonal snow: a case study over North China with field campaign measurements, *Atmos. Chem. Phys.*, 14, 11475-11491, 2014.