Observations and model simulations of snow albedo reduction in seasonal snow due to insoluble light-absorbing particles during 2014 Chinese survey

Xin Wang[1], Wei Pu[1], Yong Ren[1], Xuelei Zhang[2], Xueying Zhang[1], Jinsen Shi[1], Hongchun Jin[1],

Mingkai Dai[1], Quanliang Chen[3]

[1] Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, College of Atmospheric Sciences, Lanzhou University, Lanzhou, 730000, China
[2] Key Laboratory of Wetland Ecology and Environment, Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences, Changchun 130102, China
[3] College of Atmospheric Science, Chengdu University of Information Technology, and Plateau Atmospheric and Environment Laboratory of Sichuan Province, Chengdu 610225, China

Correspondence to: X. Wang (wxin@lzu.edu.cn)

Abstract.

A snow survey was carried out to collect 13 surface snow samples (10 for fresh snow, and 3 for aged snow), and 79 sub-surface snow samples in seasonal snow at 13 sites in January 2014 across northeastern China. A spectrophotometer and the chemical

5   analysis were used to separate snow particulate absorption by insoluble light-absorbing particles (ILAPs, e.g. black carbon, BC; mineral dust, MD; and organic carbon, OC) in snow, and the snow albedo was measured by using a field spectroradiometer during this period. In this study, a new radiative transfer model (Spectral Albedo Model for Dirty Snow, or SAMDS) is developed to simulate the

10   spectral albedo of snow based on the asymptotic radiative transfer theory. The comparison between SAMDS and the Snow, Ice, and Aerosol Radiation (SNICAR) model indicates that the spectral albedos of pure snow derived from these models agree well, however, there is a slight tendency for the SNICAR model values to become lower than SAMDS model values when BC and MD are considered. Given

15   the measured BC, MD and OC mixing ratios of 100-5000 ng g^{-1}, 2000-6000 ng g^{-1}, and 1000-30000 ng g^{-1} in surface snow across northeastern China, the snow albedo of fresh snow at 550 nm with a snow grain optical effective radius (R_{eff}) of 100 μm is generally in a range of 0.95-0.75 by using SAMDS model. The snow albedo reduction due to spherical snow grains assumed as aged snow is graduately larger than fresh

20   snow such as fractal snow grains, and hexagonal plates/colums snow grains associated with the increased BC in snow. For typically BC mixing ratios of 100 ng g^{-1} in remote areas and 3000 ng g^{-1} in heavy industrial areas across northern China, the snow albedo for internal mixing of BC and snow is lower by 0.005 and 0.036 than that of external mixing for hexagonal plates/columns snow grains with R_{eff} of 100 μm. The result also

25   shows that the simulated snow albedos by both SAMDS and SNICAR agree well with

the observed values at low ILAPs mixing ratios, but tend to be higher compared with

surface observations at high ILAPs mixing ratios.

5   **1   Introduction**

Mineral dust (MD), black carbon (BC) and organic carbon (OC) are three insoluble

light-absorbing particles (ILAPs) that play key roles in regional and global climate

(Bond et a., 2013; Dang et al., 2014; Flanner et al., 2007, 2009; Hansen et al., 2005;

IPCC, 2013; Li et al., 2016; McConnell, 2007). The MD particles can influence air

10   quality and human health through emission, transport, removal, and deposit processes

(Aleksandropoulou et al., 2011; Chen et al., 2013; Huang et al., 2014, 2015a, 2015b;

Kim et al., 2009; Li et al., 2009; Mahowald and Luo, 2003; Zhang et al., 2005). They

also act as cloud condensation nuclei that affect cloud formation (Coz et al., 2010;

Givati and Rosenfeld, 2004; Rosenfeld et al., 2001) and deliver various trace nutrients

15   to terrestrial and marine ecosystems (Acosta et al., 2011; Mahowald et al., 2005; Qiao

et al., 2013). The Taklimakan and Gobi desert, and several other deserts are well

known to be major dust sources across northern China, and MD particles can be lifted

in the atmosphere and transported to the downwelling regions via wet and dry

depositions (Chen et al., 2013; Huang et al., 2008; Jaffe, et al., 1999; Wang et al.,

20   2008; Zhang et al., 2003). But recent studies indicated that the anthropogenic dust

emissions from disrupted soils by human activities, such as deforestation, overgrazing,

agricultural and industrial activities, are not well constrained (Aleksandropoulou et al.,

2011; Tegen and Fung, 1995; Tegen et al., 2002, 2004; Thompson et al., 1988), which

differs from natural MD originating from desert regions (Che et al., 2011, 2013,

2015a; Goudie and Middleton, 2001; Li et al., 2012; Park and Park, 2014; Pu et al., 2015; Wallach and Fischer, 1970; Wang et al., 2008, 2010b). For instance, Kamani et al. (2015) indicated that MD influenced by anthropogenic activities has already been enriched with heavy metals and other toxic elements. This assumption is consistent with the results of a recent study by Huang et al. (2015a), who found that anthropogenic dust contributions to regional emissions in eastern China are 91.8%, and 76.1% in India. This may be due to larger population densities, which are characterized by more intense human activity in eastern China and India (Guan et al., 2016).

Compared to MD, carbonaceous aerosols, such as BC and OC, generated from the incomplete combustion of fossil fuels and from biomass burning are also major anthropogenic pollutants. ILAPs deposited on snow have been found to shorten the snow cover season by decreasing the snow albedo and accelerating snow melt (Flanner et al., 2007, 2009). It is widely known that small snow albedo changes can have significant effects on global warming patterns, involving changes in snow morphology, sublimation, and melt rates (Brandt et al., 2011; Hadley and Kirchstetter, 2012; Warren and Wiscombe, 1985). Warren and Wiscombe (1980) found that a mixing ratio of 10 ng g^{-1} of soot in snow can reduce snow albedo levels by 1%. Light et al. (1998) determined that 150 ng g^{-1} of BC embedded in sea ice can reduce ice albedo levels by a maximum of 30%. 1 ng g^{-1} of BC has approximately the same effect on the albedo of snow and ice at 500 nm as 50 ng g^{-1} of dust (Warren, 1982). Doherty et al. (2013) analyzed field measurements of vertical distributions of BC and other ILAPs in snow in the Arctic during the melt season and found significant melt amplification due to an increased mixing ratio of BC by up to a factor of 5. Yasunari et al. (2015) suggested that the existence of snow darkening effect in the Earth system

associated with ILAPs contributes significantly to enhanced surface warming over continents in northern hemisphere midlatitudes during boreal spring, raising the surface skin temperature by approximately 3–6 K near the snowline. Warren and Wiscombe (1985) pointed out that modeling soot in snow as an "external mixture" (impurities particles separated from ice particles) may underestimate the true effect of the impurities as a given reduction of albedo by about half as much soot, if the soot is instead located inside the ice grains as an "internal mixture". Hansen et al. (2004) and Cappa et al. (2012) noted that for a given BC mass on snow albedo, the internal mixing of BC in snow is a better approximation than external mixing, whereas internal mixing increases the BC absorption coefficient by a factor of two, for better agreement with empirical data. Hadley and Kirchstetter (2012) also indicated that increasing the size of snow grains could decrease snow albedo and amplify the radiative perturbation of BC. For a snow grain optical effective radius (R_{eff}) of 100 μm, the albedo reduction caused by 100 ng g^{-1} of BC is 0.019 for spherical snow grains but only 0.012 for equidimensional nonspherical snow grains (Dang et al., 2016). Fierce et al. (2016) pointed out that BC coated with non-absorbing particles absorbs more strongly than the same amount of BC in an uncoated particle, but the magnitude of this absorption enhancement is still a challenge. He et al. (2014) indicated 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 using a global chemical transport model in conjunction with a stochastic snow model and a radiative transfer model.

Climate models have indicated that the reduction in surface albedo caused by BC leads to global warming and nearly global melting of snow and ice (Hansen and

Nazarenko, 2004). Recent modeling studies have estimated regional and global radiative forcing caused by ILAPs deposited on snow by reducing the surface albedo (Flanner, 2013; Flanner et al., 2007; Hansen and Nazarenko, 2004; Jacobson, 2004; Koch et al., 2009; Qian et al., 2015; Zhao et al., 2014). Flanner et al. (2012) demonstrated that the simulated global-mean fraction of BC residing within ice grains of surface snow increases the global BC/snow radiative forcing by 34–86% relative to paired control simulations that apply external optical properties to all BC. Bond et al. (2013) estimated the industrial-era climate forcing of BC through all forcing mechanisms to be approximately +1.1 W m^{-2}, with 90% uncertainty bounds of +0.17 to +2.1 W m^{-2}. This value includes the net effect of BC on radiation, clouds, and snow albedo, which have been found to heavily impact the earth's climate (Bond et al, 2013; IPCC, 2013; Li et al., 2016).

Although MD is a less efficient absorber than BC and OC, field campaigns on collecting seasonal snow samples across northern China and the Himalayas have shown high MD loadings (Guan et al., 2015; Kang et al., 2016; Wang et al., 2012; Wang et al., 2013a). In some regions, especially areas with thin and patchy snow cover and mountainous regions, soil dust significantly decreases the snow albedo, exceeding the influence of BC. However, models did not capture these potentially large sources of local dust in snowpack and may overestimate BC forcing processes (Painter et al., 2007, 2010, 2012). Recently, several seasonal snow collection campaigns were performed across northern China, the Himalayas, North America, Greenland and the Arctic (Cong et al., 2015; Dang and Hegg, 2014; Doherty et al., 2010, 2014; Huang et al., 2011; Xu et al., 2009, 2012; Zhao et al., 2014). However, determining the effects of ILAPs on snow albedo reduction continues to present

numerous challenges (Huang et al., 2011; Wang et al., 2013a, 2015; Ye et al., 2012; Zhang et al., 2013a).

To our knowledge, there are only a few studies that compare modeled and observed snow albedo reduction due to ILAPs in snow (Dang et al., 2015; Flanner et al., 2007, 2012; Grenfell et al., 1994; Liou et al., 2014; Warren et al., 1980). In this study, a 2014 snow survey was performed across northeastern China to analyze light absorption of ILAPs in seasonal snow, and the comparison of snow albedo reduction due to internal/external mixed BC in snow and different snow grain shapes. In section 2, we present the experimental procedures, including a new radiative transfer model (Spectral Albedo Model for Dirty Snow, or SAMDS). After describing our methods (Sect. 2), we demonstrate the light absorption by snowpack containing ILAPs across northeastern China for less snow fallen year through a Chinese survey in 2014 following the snow surveys held in 2010 and 2012 across northern China carried out by Huang et al. (2011) and Ye et al. (2012). Then, a comparison of the snow albedo reduction under clear sky conditions measured by using a field spectroradiometer and simulated by the Snow, Ice, and Aerosol Radiation (SNICAR) model and SAMDS model based on two-stream radiative transfer solution is present. The SAMDS model is also used for the computation of light absorption by complex ILAPs in snow for application to analyze the effects of snow grain shapes (fractal grains, hexagonal plates/columns, and spheres) and internal/external mixing of BC and snow on snow albedo. Finally, conclusions are given in section 4.

2   Experimental procedures

2.1   Snow field campaign in January 2014

In 2014, there was less snowfall in January than in previous years (e.g., 2010), and only 92 snow samples (13 surface snow, and 79 sub-surface snow samples) at 13 sites were collected during this snow survey. There were 10 fresh snow, and 3 aged snow for surface snow. The snow sampling sites in this study were began at 90 (see Figure 1 and Table 1), which are numbered in chronological order followed by Wang et al. (2013a), and Ye et al. (2012). Samples from sites 90-93 were collected from grassland and cropland areas in Inner Mongolia. Sites 94-98 and sites 99-102 were located in the Heilongjiang and Jilin provinces, respectively, which are the most heavily polluted areas in northern China during winter. The snow sampling routes were similar to those used in the previous survey conducted in 2010 across northern China (Huang et al., 2011). To prevent contamination, the sampling sites were positioned 50 km from cities and at least 1 km upwind of approach roads or railways; the only exception was site 101, which was positioned downwind and close to villages. Two vertical profiles of snow samples ("left" and "right") were collected through the whole depth of the snowpack at all sites to reduce the possible contamination by artificial effects during the sampling process, and we note that the dusty or polluted layers were separately collected during the sampling process. All of the datasets in seasonal snow from Table 1 are the average values from the two adjacent snow samples through the whole depth of the snowpack. Snow grain sizes (R_m) were measured by visual inspection on millimeter-gridded sheets viewed through a magnifying glass. The snow samples were kept frozen until the filtration process was initiated. In a temporary lab based in a hotel, we quickly melted the snow samples in a microwave, let them settle for 3-5 minutes, and then filtered the resulting water samples through a 0.4-μm nuclepore filter to extract particulates.

2.2   Chemical speciation

The major water-soluble ions and trace elements in surface snow samples during this snow survey have already been investigated by Wang et al. (2015). However, the ILAPs in seasonal snow during this survey have not shown yet. For the importance of the ILAPs in snow, we will present the contribution and discuss the potential

5   emission sources of the ILAPs together with suites of other corresponding chemical constituents in seasonal snow. For instance, Hegg et al. (2009, 2010) analysed the source attribution of the ILAPs in arctic snow by using a positive matrix factorization (PMF) model consisted with trajectory analysis and satellite fire maps. Briefly, major ions (SO_4^{2-}, NO_3^-, Cl^-, Na^+, K^+, and NH_4^+) were analyzed with an ion chromatograph

10   (Dionex, Sunnyvale, CA), and trace elements of Fe and Al were measured by inductively coupled plasma mass spectrometry (ICP-MS). These analytical procedures have been described elsewhere (Yesubabu et al., 2014). In this paper, the major ions are used to retrieve the sea salt and biosmoke potassium ($K_{Biosmoke}^+$), which datasets were not shown by Wang et al. (2015). However, only SO_4^{2-}, NO_3^-, and NH_4^+ were

15   reprinted from Wang et al. (2015) to reveal the mass contribution of the ILAPs and the chemical constituents in seasonal snow during this snow survey. Previous studies have revealed considerable variations in iron (Fe) of 2-5% in dust (Lafon et al., 2006), although Al is more stable than Fe in the earth's crust. Hence, we retrieved the mass concentration of MD via the Al concentration assuming a fraction of 7% in MD

20   (Arhami et al., 2006; Lorenz et al., 2006; Zhang et al., 2003). Sea salt was estimated following the method presented in Pio et al. (2007):

$$\text{Sea salt} = Na_{Ss}^+ + Cl^- + 0.12Na_{Ss}^+ + 0.038Na_{Ss}^+ + 0.038Na_{Ss}^+ + 0.25Na_{Ss}^+, \quad (1)$$

where subscript Ss means sea salt sources, Na_{Ss} was calculated using the following formula (Hsu et al., 2009):

$$Na_{Ss} = Na_{Total} - Al \times (Na/Al)_{Crust}. \tag{2}$$

Following Hsu et al. (2009), the contribution of $K^+_{Biosmoke}$ was determined using the following equations:

$$K^+_{Biosmoke} = K_{Total} - K_{Dust} - K_{Ss}, \tag{3}$$

$$K_{Dust} = Al \times (K/Al)_{Crust}, \tag{4}$$

$$Na_{Ss} = Na_{Total} - Al \times (Na/Al)_{Crust}, \tag{5}$$

$$K_{Ss} = Na_{Ss} \times 0.038, \tag{6}$$

where $K^+_{Biosmoke}$, K_{Dust}, and K_{Ss} refer to biosmoke potassium, dust-derived potassium, and sea-salt-derived potassium, respectively. Equations (4), (5), and (6) were derived from Hsu et al. (2009) and Pio et al. (2007).

2.3 Spectrophotometric analysis

Recent studies indicated that the light absorption by MD should be more sensitive to the presence of strongly absorbing iron oxides such as hematite and goethite than to other minerals (Alfaro et al., 2004; Sokolik and Toon, 1999). Thus it is now possible to assess the absorption properties of MD by using iron oxide content (Bond et al., 1999). In this study, the iron (Hereinafter simply "Fe") in seasonal snow is assumed to be originating from MD during this survey, which followed the procedures performed by Wang et al., (2013a). We measured the mixing ratios of BC and OC using an integrating sphere/integrating sandwich spectrophotometer (ISSW), which was first described by Grenfell et al. (2011) and used by Doherty et al. (2010, 2014) and Wang et al. (2013a). The equivalent BC (C^{equiv}_{BC}), maximum BC (C^{max}_{BC}), estimated BC (C^{est}_{BC}), fraction of light absorption by non-BC ILAPs (f^{est}_{non-BC}), and absorption Ångstrom exponent of all ILAPs ($Å_{tot}$) were described by Doherty et al. (2010). Previous studies

have concluded that ILAPs are primarily derived from BC, OC, and Fe. The mass loadings of BC (L_{BC}) and OC (L_{OC}) were calculated using the following equation:

$$\tau_{tot}(\lambda) = \beta_{BC}(\lambda) \times L_{BC} + \beta_{OC}(\lambda) \times L_{OC} + \beta_{Fe}(\lambda) \times L_{Fe}. \tag{7}$$

Here, $\tau_{tot}(\lambda)$ refers to the measured optical depth. L_{BC} and L_{OC} can be determined from this equation assuming that the mass absorption coefficients (MACs) for BC, OC, and Fe are 6.3, 0.3, and 0.9 $m^2 g^{-1}$, respectively, at 550 nm and that the absorption Ångstrom exponents (Å or AAE) for BC, OC and Fe are 1.1, 6, and 3, respectively (e.g., Equations (2) and (3) in Wang et al., 2013a).

2.4   Aerosol optical depth and snow albedo measurements

The Microtops Π Sun photometer has been widely used to measure aerosol optical depth (AOD) in recent years (More et al., 2013; Porter et al., 2001; Zawadzka et al., 2014), and is recognized as a very useful tool for validating aerosol retrievals from satellite sensors. Ichoku et al. (2002b) and Morys et al. (2001) provided a general description of the Microtops Π Sun photometer's design, calibration, and performance. To better understand the background weather conditions in the local atmosphere during this snow survey, we used a portable and reliable Microtops Π Sun photometer at wavelengths of 340, 440, 675, 870, and 936 nm instead of the CE318 sun tracking photometer to measure the surface AOD in this study. AOD measurements were collected in cloud-free conditions between 11:00 am and 1:00 pm (Beijing local time) to prevent the effects of optical distortions due to large solar zenith angles. Then, the Moderate Resolution Imaging Spectroradiometer (MODIS) on the Aqua and Terra satellites was used to retrieve the AOD and fire spot datasets (Kaufman et al., 1997; Zhang et al., 2013b; Zhao et al., 2014). The retrieved MODIS AOD is reliable and accurate when applied to three visible channels over vegetated land and ocean

surfaces (Chu et al., 2002; Ichoku et al., 2002a; Remer et al., 2002). Fire locations are based on data provided by the MODIS FIRMS system from October 2013 to January 2014. The land-cover types (Figures 6 and 7) were obtained from the Collection 5.1 MODIS global land-cover type product (MCD12C1) at a 0.05° spatial resolution and

5   included 17 different surface vegetation types (Friedl et al., 2010; Loveland and Belward, 1997).

Snow albedo plays a key role in affecting the energy balance and climate in the cryosphere (e.g. Hadley and Kirchstetter, 2012; Liou et al., 2014; Warren and Wiscombe, 1985). Wright et al. (2014) indicated that the spectral albedo measured by

10   using an Analytical Spectral Devices (ASD) spectroradiometer at 350-2200 nm is in agreement with albedo measurements at the baseline Surface Radiation Network (BSRN). Wuttke et al. (2006a) pointed out that the spectroradiometer instrument is considered as more capable, rapid, and mobile to conduct spectral albedo measurements during short time periods, especially in the very cold regions (e.g. in

15   the Arctic). The major advantage is the more extensive wavelength range, and the cosine error is less than 5% for solar zenith angles below 85° at the wavelength of 320 nm (Wuttke et al., 2006a, b). In this study, snow albedo measurements were obtained using a HR-1024 field spectroradiometer (SVC, Spectra Vista Corporation, Poughkeepsie, NY, USA). This instrument has a spectral range of 350-2500 nm with

20   resolutions of 3.5 nm (350-1000 nm), 9.5 nm (1000-1850 nm), and 6.5 nm (1850-2500 nm). Normally, the relative position of the spectroradiometer is at a distance of 1m from the optical element for the active field of view (Carmagnola et al., 2013). A standard "white" reflectance panel with a VIS–SWIR broadband albedo of 0.98 (P_λ) was used to measure the reflectance spectra along with the target. The

25   reflectance spectra of surface snow (R_s) and the standard panel (R_p) were measured at

least ten times. Then, the snow albedo (α) was calculated as follows:

$$\alpha = (R_s/R_p) \times P_\lambda. \tag{8}$$

The nominal filed of view (FOV) lens is 8° to enable the instrument to look at different size targets. In order to receive more direct solar radiation, the direction of the instrument was oriented to the Sun Horizon angles to measure snow albedo. 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. Further information on HR-1024 field spectroradiometer user and on the calibration procedure can be found in Wright et al. (2014). The measured solar zenith angles and the other parameters used to simulate snow albedo in this study have been labeled in Figure 11.

2.5   Model simulations

BC and MD sensitivity effects on the snow albedo simulated by the Snow, Ice, and Aerosol Radiation (SNICAR) model have been validated through recent simulations and field measurements (Flanner et al., 2007, 2009; Qian et al., 2014; Zhao et al., 2014). We used the offline SNICAR model to simulate the reduction in surface snow albedo resulting from ILAPs contamination (Flanner et al., 2007), and we compared the results with our spectroradiometer surface measurements. The SNICAR model calculates the snow albedo as the ratio of the upward and downward solar flux at the snow surface. The measured parameters, including the measured snow grain radius (R_m), snow density, snow thickness, solar zenith angle θ, and mixing ratios of BC and

MD, were used to run the SNICAR model under clear sky conditions. The visible and near-infrared albedos of the underlying ground were 0.2 and 0.4, respectively, as derived from MODIS remote sensing. The MAC of BC was assumed to be 7.5 m^2 g^{-1} at 550 nm.

5   The Spectral Albedo Model for Dirty Snow (SAMDS) for calculating spectral snow albedo as a function of the snow grain radius, the mixing ratios of ILAPs (BC, MD, and OC), and MACs of impurities was used and is based on asymptotic radiative transfer theory. A detailed description of asymptotic analytical radiative transfer theory for SAMDS could be found by Zhang et al. (2016). Briefly, the surface albedo

10   can be calculated using the following asymptotic approximate analytical solution derived from radiative transfer theory (Kokhanovsk and Zege, 2004; Rozenberg, 1962; Zege et al., 1991):

$$R_d(\lambda) = \exp(-4S(\lambda)\mu(\nu_0)). \qquad (9)$$

Here, $R_d(\lambda)$ is the plane albedo, ν_0 is the solar zenith angle, and $\mu(\nu_0)$ refers to the

15   escape function in radiative transfer theory, and can be parameterized following Kokhanovsky and Zege (2004):

$$\mu(\nu_0) = \frac{3}{7}(1+2\cos\nu_0), \qquad (10)$$

where λ is the wavelength, $S(\lambda)$ is the similarity parameter, and

$$S(\lambda) = \sqrt{\frac{\sigma_{abs}}{3\sigma_{ext}(1-g)}}. \qquad (11)$$

20   Here, σ_{abs} and σ_{ext} are the absorption and extinction coefficients, respectively, and g is the asymmetry parameter (the average cosine of the phase function of the medium). According to Equations (18) and (25) in Kokhanovsky and Zege (2004), the extinction coefficients of particles can be expressed as follows:

$$\sigma_{ext} = \frac{l_{tr}}{1-g} = \frac{3C_v}{2r_{eff}}, \qquad (12)$$

where l_{tr} is the photon transport path length, C_v is the volumetric snow particle concentration, and r_{eff} is the effective grain size, which is equal to the radius of the volume-to-surface equivalent sphere: $r_{eff} = \frac{3\overline{V}}{4\overline{A}}$, where $\overline{V}$ and $\overline{A}$ are the average volume and average cross-sectional (geometric shadow) area of snow grains, respectively.

The absorption coefficient σ_{abs} in Equation (11) for arbitrarily shaped and weakly absorbing large grains is proportional to the volume concentration (Kokhanovsky and Zege, 2004):

$$\sigma_{abs} = B \cdot \frac{4\pi k(\lambda)}{\lambda} \cdot C_v, \tag{13}$$

where $k(\lambda)$ is the imaginary component of the complex refractive index for ice, and B is a factor that is only dependent on the particle shape. The theory based on the ray-optics approach shows that g in Equation (11) and B in Equation (13) are 0.89 and 1.27 for spheres, 0.84 and 1.50 for hexagonal plates/columns, and 0.75 and 1.84 for fractal grains, respectively.

The total absorption coefficient, σ_{abs}, can be derived from the absorption by snow, σ_{abs}^{snow}, and the absorption by ILAPs (σ_{abs}^{BC}, σ_{abs}^{MD}, and σ_{abs}^{OC}):

$$\sigma_{abs} = \sigma_{abs}^{snow} + \sigma_{abs}^{BC} + \sigma_{abs}^{MD} + \sigma_{abs}^{OC}. \tag{14}$$

The hemispherical reflectance with a zenith angle ν_0 can be expressed as follows:

$$R_d(\lambda) = \exp\left(-4 \cdot \sqrt{\frac{4B}{9(1-g)} \cdot \frac{2\pi \cdot r_{eff}}{\lambda} \cdot k(\lambda) + \frac{\rho_{ice} \cdot 2r_{eff}}{9(1-g)} \cdot MAC_{abs}^{BC} \cdot C_{BC}^* + \frac{\rho_{ice} \cdot 2r_{eff}}{9(1-g)} \cdot MAC_{abs}^{MD} \cdot C_{MD}^* + \frac{\rho_{ice} \cdot 2r_{eff}}{9(1-g)} \cdot MAC_{abs}^{OC} \cdot C_{OC}^*} \cdot \frac{3}{7}(1+2\cos\nu_0)\right), \quad \text{for spherical grains;}$$

$$\exp\left(-\sqrt{94.746 \cdot \frac{r_{eff}}{\lambda} \cdot k(\lambda) + 5.163 \cdot r_{eff} \cdot (MAC_{abs}^{BC} \cdot C_{BC}^* + MAC_{abs}^{MD} \cdot C_{MD}^* + MAC_{abs}^{OC} \cdot C_{OC}^*) \cdot (1+2\cos\nu_0)}\right), \quad \text{for hexagonal grains;}$$

$$= \exp\left(-4.95 \cdot \sqrt{\frac{\pi \cdot r_{eff} \cdot (k(\lambda) + \alpha \cdot C_{BC}^* + \beta \cdot C_{MD}^* + \chi \cdot C_{OC}^*)}{\lambda} \cdot (1+2\cos\nu_0)}\right), \quad \text{for hexagonal grains;} \qquad (15)$$

$$\exp\left(-4.38 \cdot \sqrt{\frac{\pi \cdot r_{eff} \cdot (k(\lambda) + \alpha \cdot C_{BC}^* + \beta \cdot C_{MD}^* + \chi \cdot C_{OC}^*)}{\lambda} \cdot (1+2\cos\nu_0)}\right), \quad \text{for fractal grains.}$$

Previous studies have also shown that the spectral snow albedo is more sensitive to snow grain size and light conditions than BC contamination and snow depths at near-infrared wavelengths (Warren, 1982). Therefore, the snow grain optical effective radius (R_{eff}) was retrieved based on the spectral albedo measured at $\lambda=1.3$ µm, where

5   snow grain size dominates the snow albedo variations and the effects of ILAPs at this wavelength are negligible (Warren and Wiscombe, 1980).

3   Results

3.1   The spatial distribution of AOD

The AOD is a major optical parameter for aerosol particles and a key factor affecting

10   global climate (Holben et al., 1991, 2001, 2006; Smith et al., 2014; Srivastava and Bhardwaj, 2014). Most of the snow samples were collected in the afternoon at the Aqua-MODIS (13:30 LT) overpass time in order to compare the local AODs in sampling sites by using an spectroradiometer with the satellite remote sensing. The AOD spatial distribution derived from the Aqua-MODIS satellite over northern China

15   associated with sampling site numbers is shown in Figure 1 during this snow survey. The average AOD in the studied area ranged from 0.1 to 1.0 and exhibited strong spatial inhomogeneity. The largest AOD (up to approximately 1.0) retrieved from the MODIS satellite were associated with anthropogenic pollution over northeastern China during the 2014 sampling period. These large values, which exceeded 0.6, were

20   related to local air pollution from industrial areas (Che et al., 2015b; Wang et al., 2010a). In contrast, the MODIS-Aqua results indicate that the smallest AOD (as low as 0.1) at 550 nm were found over the Gobi Desert in Inner Mongolia and were related to strong winter winds. Similar patterns in the retrieved MODIS AOD were found by Zhao et al. (2014) and Zhang et al. (2013b). Although previous studies have

indicated that AOD across northeastern China are among the highest in East Asia (Ax et al., 1970; Bi et al., 2014; Che et al., 2009; Routray et al., 2013; Wang et al., 2013b; Xia et al., 2005, 2007), field experiments of aerosol optical properties across northeastern China have been limited. Compared with the retrieved AOD by the remote sensing, the surface measurements of AOD were also conducted during this snow survey. Generally, the measured AOD were gradually higher from Inner Mongolia regions moved to the industrial area across northeastern China. In Inner Mongolia, the average AOD was less than 0.25 for sites 90 to 93 under clear sky conditions. We found large discrepancy of 40-50% in the same area within the 1-hour measurements collected from sites 95a and 95b, which could be possibly correlated with regional biomass burning. However, AOD exceeded 0.3 at sites 98 and 101, which were significantly influenced by anthropogenic air pollution from industrial areas across northern China. MODIS active fires are often spatially distributed over northeastern China and mainly result from human activities during cold seasons.

3.2   Contributions to light absorption by ILAPs

Generally, we only collected the aged surface snow in three sites, while fresh surface snow was collected in the other sites during this snow survey in January 2014. C_{BC}^{est}, C_{BC}^{max}, C_{BC}^{equiv}, f_{non-BC}^{est}, Å$_{tot}$ and snow parameters, such as snow depth, snow density, measured snow grain radius (R_m), and snow temperature, are given in Table 1 and Figure 3 for each snow layer following Wang et al. (2013a). 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. The snow samples were collected from drifted snow in Inner Mongolia, and the mass loadings of ILAPs in seasonal snow are

mainly due to blowing soil dust. Therefore, the vertical profiles of snow samples

mixed with blowing soil from these sites are insufficient to represent the seasonal

evolution of wet and dry deposition to snow (Wang et al., 2013a). However, the light

absorption of ILAPs is still dominated by OC in these regions, which has been

5   illustrated in the following section. 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 in previous studies because of snow

10   melting by solar radiation and the ILAPs (Hadley and Kirchstetter, 2012; Motoyoshi

et al., 2005; Painter et al., 2013; Pedersen et al., 2015). The snow density exhibited

little geographical variation across northern China at 0.13 to 0.38 g cm^{-3}. High snow

densities result from melting or snow aging. Similar snow densities have been found

in the Xinjiang region in northern China (Ye et al., 2012). At site 90, we only

15   collected one layer of snow samples from central Inner Mongolia, and C_{BC}^{est} was 330

ng g^{-1} for aged snow. Along the northern Chinese border at sites 91-95, C_{BC}^{est} in the

cleanest snow ranged from 30 to 260 ng g^{-1}; only a few values exceeded 200 ng g^{-1}.

The $f_{non\text{-}BC}^{est}$ value varied remarkably from 29 to 78%, although BC was still a major

absorber in this region. Heavily polluted sites were located in industrial regions across

20   northeastern China (sites 99-102). The surface snow C_{BC}^{est} in this region ranged from

510 to 3700 ng g^{-1}, and the highest C_{BC}^{est} in the sub-surface layer of the four sites was

2900 ng g^{-1} in Table 1. In addition, $f_{non\text{-}BC}^{est}$ was typically 35-74%, indicating

significant light-absorbing contributions by OC and MD from human activity in the

heavily polluted areas. Å_{tot} ranged from 2.1 to 4.8. A higher Å_{tot} is a good indicator of

soil dust, which is primarily driven by the composition of mineral or soil dust. In contrast, a lower $Å_{tot}$ of 0.8-1.2 indicates that ILAPs in the snow are dominated by BC (Bergstrom et al., 2002; Bond et al., 1999).

To better understand the distribution of C_{BC}^{est} in seasonal snow across northern China, the spatial distribution of C_{BC}^{est} in the surface and average snow measured during this snow survey are shown in Figure 4. The spatial distributions of C_{BC}^{est} in the surface and average snow measured using the ISSW spectrophotometer during the 2014 survey generally ranged from 50 to 3700 ng g^{-1} and 60 to 1600 ng g^{-1}, with the medium values of 260 ng g^{-1}, and 260 ng g^{-1}, respectively. These variations of C_{BC}^{est} were very similar to those of the previous snow campaign by Wang et al. (2013a), however, much higher than those in the Xinjiang region of northwestern China (Ye et al., 2012), along the southern edge of the Tibetan Plateau (Cong et al., 2015), and across North America (Doherty et al., 2014).

Figure 5 compares C_{BC}^{est} values measured via the ISSW method with the calculations during 2010 (Wang et al., 2013a) and 2014 snow surveys. Ideally, C_{BC}^{est} calculated using Equation (7) should be equal to that measured via the ISSW method. The two results agreed very well (R^2=0.99), indicating that Equation (7) agreed well for this measurement in Figure 5. Thus, the C_{BC}^{est} measured via the ISSW method was found to be reasonable and reliable. To compare with the mixing ratio of OC calculated from Equation (7), we used the calculated C_{BC}^{est} listed in Figures 6-7 and Table 2 in the following sections.

In Figure 6, the sampling areas were located in grasslands, croplands, and urban and built-up regions across northern China that are likely influenced by human activity (Huang et al., 2015a). According to Table 2 and Figure 6, the NH_4^+ concentrations emitted from agricultural sources at all sites accounted for less than 2.8% because the

sites were positioned 50 km from cities. However, large fractions of both SO_4^{2-}, and

NO_3^- were observed, varying from 14.8 to 42.8% at all sites, with the highest fraction

of 24.2-42.8% found in industrial areas. These results show that SO_4^{2-}, and NO_3^-

made the greatest contributions to the total chemical concentration in the surface snow

5   and are significant anthropogenic emissions of fossil-fuel combustion in heavy

industrial areas. More specifically, the largest MD contribution ranged from 35.3 to

46% at sites 91 to 95 due to strong winds during winter; the MD fractions were only

5.7 to 31% at the other sites. Fractions of BC and OC were similar to those above,

showing that biomass burning was a major source during winter in the sampling

10   region. Zhang et al. (2013b) showed that OC and BC fractions vary more widely in

the winter than other seasons due to industrial activity in China. Sulfate peaks were

found in summer (15.4%), whereas nitrate peaks were observed in spring (11.1%).

$K_{Biosmoke}^+$ was found to be a good tracer of biomass burning, ranging from 1.3 to 5.1%

along the northern Chinese border compared to lower values found at lower latitudes

15   (1.5-2.3%), and it exhibited much higher contributions in Inner Mongolia and along

the northern Chinese border due to increased emissions from cooking, open fires, and

agricultural activities. The fraction of sea salt was found to range from 6.3 to 20.9%.

Wang et al. (2015) showed that higher Cl^-/Na^+ ratios in seasonal snow found in the

2014 Chinese survey were 1-2 times greater than those of seawater, implying that they

20   constituted a significant source of anthropogenic Cl^-.

As described by Wang et al. (2013a), light absorption by ILAPs can be determined

from ISSW measurements combined with a chemical analysis of Fe concentrations by

assuming that the light absorption of dust is dominated by Fe; however, Fe can also

originate from industrial emissions, such as the metal and steel industries (Hegg et al.,

2010; Ofosu et al., 2012). Doherty et al. (2014) used a similar method to distinguish contributions of ILAPs in snow in central North America. Although heavy MD loading was observed in 2014 snow survey, the fraction of light absorption due to MD (assumed to exist as goethite) was generally less than 10% across northeastern China (Figure 7), which was much smaller than that observed in the Qilian Mountains (e.g., Figure 11 of Wang et al., 2013a). Here, light absorption was mainly dominated by BC and OC in snow in January 2014. By contrast, the fraction of light absorption due to BC varied from 48.3 to 88.3% at all sites, with only one site dominated by OC (site 90 in central Inner Mongolia). Compared to the light absorption patterns in the Qilian Mountains, MD played a less significant role in particulate light absorption in snow across the northeastern China sampling area.

3.3   Simulations of snow albedo

Snow albedo reduction due to BC has been examined in previous studies (Brandt et al., 2011; Hadley and Kirchstetter, 2012; Yasunari et al., 2010). Here, a new radiative transfer model (SAMDS) is developed to assess the effects of ILAPs in snow, the snow grain shapes, and the internal/external mixing of BC and snow on snow albedo based on the asymptotic radiative transfer theory. The snow albedos derived from the SNICAR and SAMDS models are presented in Figure 8. We ran the models at a solar zenith angle θ of 60°, which is comparable with our experimental method for measuring snow albedo across northeastern China. The BC MAC used in the two models was 7.5 m^2 g^{-1} at 550 nm, which was assumed in the most recent climate assessment and is appropriate for freshly emitted BC (Bond and Bergstrom, 2006; Bond et al., 2013; Warren, 1982). Mixing ratios of BC, MD, and OC were chosen to exhibit the following ranges: 0.1-5 μg g^{-1}, 2-6 μg g^{-1}, and 1-30 μg g^{-1}, respectively,

encompassing the values measured in snow surfaces across northeastern China in this study and in previous research (Doherty et al., 2010, 2014; Wang et al., 2013a, 2014; Warren and Wiscombe, 1980; Ye et al., 2012). Results showed that the albedo of fresh snow at 550 nm with R_{eff} of 100 μm simulated by SAMDS is generally in a range of 0.95-0.75 for ILAPs-contaminated snow across northeastern China (Figure 8). The spectral albedos of pure snow derived from the SNICAR (dash lines) and SAMDS (solid lines) models agree well. However, there is a slight tendency for the SNICAR model values to become lower than SAMDS model values when BC and MD are considered. The 1-3% deviation between the SNICAR and SAMDS modeled snow albedos at 550 nm for BC mixing ratios of 1-5 μg g^{-1} indicates that albedo reduction by ILAPs in the SNICAR model was greater than that in the SAMDS model. This deviation is in part due to the different parameterization of snow grain shapes, mixing states of snow and BC, and physical-chemical properties of impurities between the two models (Zhang et al., 2016). More notably, snow albedos decrease significantly within the UV-visible wavelength, especially for the higher OC (dotted lines) mixing ratios in Figure 8. This may be attributed to the fact that OC strongly absorbs UV-visible radiation and masks BC absorption for high AAE of OC, which decreases remarkably with increasing wavelengths (Warren and Wiscombe, 1980).

As is shown in Figure 9, we also estimated the reduction in the spectrally weighted snow albedo for different R_{eff} values using the SNICAR and SAMDS models. A larger reduction in snow albedo by both BC and MD-contaminated snow was found for larger snow grains (Figure 9a-b). For example, snow albedo reduction attributable to 1 μg g^{-1}, 1 μg g^{-1}, and 10 μg g^{-1} for BC, MD, and OC, respectively, was 37%, 41%, and 38% greater in 200 μm snow grains (0.081, 0.0019, and 0.047) than that in 100 μm snow grains (0.059, 0.0013, and 0.034). Both the SNICAR and SAMDS models

indicated that the snow albedo is more sensitive to BC, especially at low ILAPs mixing ratios. For example, 200 ng g^{-1} of BC decreased 0.03 snow albedo at R_{eff} of 200 μm, which is much larger than the snow albedo reduction of 0.003 and 0.018 by 2000 ng g^{-1} of MD and OC at R_{eff} of 200 μm. As Hadley and Kirchstetter (2012)

5   noted, compared with pure 55 μm snow grains, 300 ng g^{-1} of BC contamination and growth of R_{eff} to 110 μm cause a net albedo reduction of 0.11 (from 0.82 to 0.71), causing snow to absorb 61% more solar energy. The difference of snow albedo reductions between SNICAR and SAMDS models increases as BC mixing ratio as well as R_{eff} increases (Figure 9a). However, the snow albedo reduction simulated by

10 SNICAR is not always larger than that by SAMDS when the input contaminant is MD instead of BC (Figure 9b). For example, for R_{eff} of 100 μm, the snow albedo reduction for SAMDS is higher than that for SNICAR at MD mixing ratio < 2600 ng g^{-1}, but lower than that for SNICAR at MD mixing ratio > 2600 ng g^{-1}. The turning point of MD mixing ratio is not constant and depends on the value of R_{eff}. The complex

15 difference may be a result of the different input optical properties of MD between SAMDS and SNICAR models (Flanner et al., 2007; Zhang et al., 2016). SAMDS model also considers the effect of OC on snow albedo, which is not included in SNICAR model. The albedo reduction by OC is nonnegligible due to its high loading. As is shown, 5000 ng g^{-1} of OC was found to reduce the snow albedo by 0.016-0.059

20 depending on the snow grain size (50-800 μm).

Previous studies have also indicated that the mixing ratio of BC (10-100 ng g^{-1}) in snow may decrease its albedo by 1-5% (Hadley and Kirchstetter, 2012; Warren and Wiscombe, 1980). Liou et al. (2011) demonstrated that a small BC particle on the order of 1 μm internally mixed with snow grains could effectively reduce visible snow

25 albedo by as much as 5-10% using a geometric-optics surface-wave approach. They

also found that internal mixing of BC in snow reduces snow albedo substantially more than external mixing, and the snow grain shape plays a critical role in snow albedo calculations through its forward scattering strength by modeling the positions of BC internally mixed with different snow grain types (Liou et al., 2014). Figure 10a illustrates the effect of snow shape (fractal grains, hexagonal plates/columns, and spheres) on snow albedo at the spectral wavelengths of 400 nm-1400 nm with R_{eff} of 100 μm simulated by SAMDS model. As is shown, the differences of snow albedo caused by three snow shapes are remarkable. The snow albedo for spherical snow grains is higher than that for the other two shapes, which is because that, the scattering by spherical snow grains is more in forward direction and less to the sides, resulting in a larger g and a smaller B as discussed in section 2.5. In addition, the snow albedo reduction for aged snow such as spherical snow grains is graduately larger than fresh snow such as fractal snow grains, and hexagonal plates/colums snow grains with the increased BC in snow. It shows that snow albedo by spherical snow grains is typically lower by 0.017-0.073 than the fractal snow grains, and by 0.008-0.036 than the hexagonal plates/columns snow grains as a function of BC mixing ratios (0-5000 ng g^{-1}). Dang et al. (2016) assessed the effects of snow grain shapes on snow albedo using the asymmetry factors g of nonspherical ice crystal developed by Fu (2007). They obtained similar result that the albedo reduction caused by 100 ng g^{-1} of BC for spherical snow grains is larger by 0.007 than nonspherical snow grains with the same area-to-mass ratio for R_{eff} of 100 μm. Figure 10b shows the spectral albedo of snow for the internal/external mixing of BC and snow with R_{eff} of 100 μm for a solar zenith angle θ of 60° as a function of BC mixing ratio. For a given shape (hexagonal plates/columns), we found that snow albedo as a function of BC mixing ratios calculated from this study decreases as the fraction of the internal mixing increases

(Figure 10b). In previous studies, the BC mixing ratios in seasonal snow were up to 3000 ng g^{-1} due to heavy industrial activities across northern China, but the lowest mixing ratios of BC were found in the remote northeastern on the border of Siberia, with a median value in surface snow of 100 ng g^{-1} (Huang et al., 2011; Wang et al., 2013a, 2014; Ye et al., 2012). As a result, snow albedo by internal mixing of BC and snow is lower than external mixing by up to 0.036 for 3000 ng g^{-1} BC in snow in the heavy industrial regions across northeastern China, whereas by low to 0.005 for 100 ng g^{-1} BC in snow in the further north China near the border of Siberia. We indicated that the snow grain shape effect on snow albedo between spherical snow grains and fractal snow grains is relatively larger than the effect of the internal/external mixing of BC and snow as a function of the BC mixing ratios. However, He et al. (2014) also pointed out that the snow albedo reductions computed by previous models under assorted assumptions vary by a factor of 2 to 5.

3.4 Comparison between the observed and modeled snow albedo

Although the snow albedo reduction due to ILAPs has been investigated by model simulations in recent studies (Brandt et al., 2011; Flanner et al., 2007; Hadley and Kirchstetter, 2012; Liou et al., 2011, 2014; Warren and Wiscombe, 1980), we note that there were still limited field campaigns on collecting snow samples and measuring ILAPs in seasonal snow associated with the snow albedo reduction at the middle latitudes in northern hemisphere (Doherty et al., 2010, 2014; Wang et al., 2013a; Ye et al., 2012). In this study, we measured the snow albedo at six sites under clear sky conditions during this snow survey, and the comparison of snow albedo reduction between surface measurements and the SNICAR and SAMDS models based on Toon et al.'s (1989) two-stream radiative transfer solution was investigated in

Figure 11. The model input parameters including θ, R_m, R_{eff}, and the mixing ratios of BC (C_{BC}^{est}), MD (C_{MD}) and OC (C_{OC}) were also displayed in Figure 11. The MAC of BC used in the ISSW is 6.3 $m^2 g^{-1}$ at 550 nm, although a value of 7.5 is used in the SNICAR and SAMDS models. Thus, the value of C_{BC}^{est} was corrected by dividing it by 1.19 (see Figure 11) when BC was used as the input parameter to the snow albedo models (Wang et al., 2013a). The snow albedos measured at 550 nm varied considerably from 0.99 to 0.61 due to different mixing ratios of ILAPs and snow parameters, such as snow grain size. The snow albedos predicted by the SNICAR and SAMDS models agreed well at each site based on the same input parameters. The snow albedos of the SNICAR and SAMDS models retrieved with R_m complemented the surface measurements for lower values of C_{BC}^{est}, C_{MD}, and C_{OC} (Figure 11a-d). The highest C_{BC}^{est} values were 1500 ng g^{-1} (corrected value of 1200 ng g^{-1}) and 3700 ng g^{-1} (corrected value of 3100 ng g^{-1}) at sites 98 and 101, respectively, across industrial regions. The OC and MD mixing ratios were as high as 32000 ng g^{-1} and 3900 ng g^{-1} in this region. There, we found a larger difference in snow albedo of up to 0.2 at higher C_{BC}^{est}, C_{MD}, and C_{OC} values between the surface measurements and the modeled albedos by both SAMDS and SNICAR models with the input of R_m (red and blue solid lines in Figure 11e-f). When the snow albedo at the inferred wavelength simulated by using R_m was not accounted for, we also calculated the snow albedos by the SAMDS and SNICAR models by using R_{eff} as shown in Figure 11 (light red and blue shaded bands). Results showed that, compared with the snow albedos simulated by using R_m, these values were more approximate to the surface measurements, especially at near-infrared wavelengths, although still a slight higher than the surface measurements (gray shaded bands). This may be attributed to the fact that R_{eff} at these two sampling sites was much larger than R_m. We innovatively suppose that, for the

same snow grains, ILAPs are able to enhance the R_{eff} in spite of the same R_m. Nevertheless, due to the limited measurements of snow albedo, this supposition is quite uncertain and needed to be verified by numerous field measured snow albedos.

Combinations of the results of Figure 9 and Figure 11 that BC, OC, and MD are three

5 types of ILAPs found in snow that can reduce spectral snow albedo levels and reduced snow albedo due to ILAPs in our measurements were generally comparable to that found in the SAMDS and commonly used SNICAR models (Flanner et al., 2007; Zhang et al., 2016) when the mixing ratios of ILAPs are not quite high, we indicate that 100-500 ng g^{-1} of BC can lower the snow albedo by 0.014-0.039 relative

10 to pure snow with a snow grain size of 100 μm according to our snow field campaign, and MD was found to be a weak absorber due to its lower MD MAC, supporting previous observations made by Warren and Wiscombe (1980). The OC MAC is also lower and comparable to that of the MD. A clear decreasing trend in the surface snow albedo due to the high ambient mixing ratios of OC from Inner Mongolia to

15 northeastern China was found. The radiative transfer modeling results presented by Zhang et al. (2016) and measurement results of this study show that the spectral albedo of snow reduction due to the increased OC mixing ratios (above 20 μg g^{-1}) is larger for a factor of 3 by assuming the snow grain size of 800 μm compared to 100 μm.

20 **4   Conclusions**

In this study, a snow survey was performed in January 2014, and 92 snow samples were collected at 13 sites across northern China. We found that higher AODs measured using a sun photometer and remote sensing devices showed that heavily polluted areas remain in industrial regions across northern China. The measured C_{BC}^{est}

through the 2014 survey via the ISSW spectrophotometer in surface and average snow of 50 to 3700 and 60 to 1600 ng g^{-1}, with the medium values of 260 ng g^{-1}, and 260 ng g^{-1}, respectively, were much larger than those of previous snow field campaigns. The chemical composition analysis showed that the mass contributions in seasonal snow was dominated by OC and MD. However, assuming the MACs for BC, OC, and Fe are 6.3, 0.3, and 0.9 m^2 g^{-1}, respectively, at 550 nm, light absorption was still dominated by BC and OC in seasonal snow during the entire campaign. The light-absorbing contribution of the MD was larger at high latitudes than at low latitudes due to strong winds transporting snow. Then, we indicated that the new spectral snow albedo model (SAMDS) can be used to investigate the snow albedo influenced by the internal/external mixing of BC and snow, irregular morphology of snow grains and the vertical distribution of snow grains and impurities for multilayer snow. For instance, the snow albedo for spherical snow grains is typically lower than that for the fractal snow grains and hexagonal plates/columns snow grains with R$_{eff}$ of 100 μm. The internal mixing of BC and snow absorbs substantially more light than external mixing. For fresh snow grains of hexagonal plates/columns with R$_{eff}$ of 100 μm, the difference of snow albedo between internal and external mixing of BC and snow is up to 0.036 for 3000 ng g^{-1} BC in snow in the heavy industrial regions across northeastern China, whereas by low to 0.005 for 100 ng g^{-1} BC in snow in the further north China near the border of Siberia. The spectral albedo of snow reduction caused by OC (20 μg g^{-1}) is larger by up to a factor of 3 for a snow grain size of 800 μm compared to 100 μm by using SAMDS model.

A comparison between measured and simulated snow albedos was conducted. Generally, the snow albedos measured from a spectroradiometer and simulated using the SNICAR and SAMDS models agreed well at the lower mixing ratios of BC, MD,

and OC. However, a large discrepancy in snow albedo between the model simulations and surface measurements for heavy loading of ILAPs in snow was found by using R_m. We demonstrate that the simulated snow albedo reduction by SMDAS and SNICAR models is significantly enhanced by using R_{eff} of snow grains compared with R_m, especially in the case of near-infrared wavelengths. Although the MAC of OC is much lower than that of BC, we found that OC was a major absorber in snow due to its high mixing ratio from human activities occurring across northeastern China. Moreover, 5000 ng g^{-1} of OC was found to reduce the snow albedo by 0.016-0.059 depending on the snow grain size and aging period. Therefore, we suggest that the mixing ratio of OC should be added as an input parameter to the SNICAR model for determining snow albedos.

Although the SAMDS model might be useful to researchers who are conducting studies involving ILAPs and snow interaction and feedback in snow albedo change, we indicate that further snow surveys across northern China should be performed for the following reasons: (1) large variations of ILAPs in seasonal snow across northern China can lead higher uncertainties of snow albedo reduction, especially in the industrial regions, and (2) we only measured the snow albedo at 6 sampling sites by using the spectroradiometer in the clear sky condition due to much less snow fallen in January 2014 than that in previous years. Comparing model simulations with the observations, we found that R_{eff} could seemingly be enhanced by the high mixing ratios of ILAPs in snow, however, we note that further snow surveys on measuring snow albedo should be conducted to reveal this phenomenon. Finally, there are large uncertainties in simulating snow albedo reduction and radiative forcing due to the ILAPs mixed with snow/ice and the irregular morphology of snow grains, the potential snow albedo change for aged snow should be investigated in the following

snow surveys accordingly to test the capability of SAMDS model, which will provide

more valuable and useful information for the climate models.

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Table 1. Statistics in seasonal snow variables measured using an ISSW in the study sites. All of the datasets are the average values from the left and right snow samples. The site numbers beginning at 90 in this study are numbered in chronological order followed by Wang et al. (2013a), and Ye et al. (2012).

Site	Layer	Latitude N	Longitude E	Snow type	Site average snow depth (cm)	Sample depth (cm) Top	Sample depth (cm) Bottom	Temperature (°C)	Snow density (g cm⁻³)	R_m (mm)	C_{BC}^{equiv} (ng g⁻¹)	C_{BC}^{max} (ng g⁻¹)	C_{BC}^{est} (ng g⁻¹)	f_{non-BC}^{est} (%)	Å_{tot} 450:600 nm
90	1	45°02'44"	116°22'45"	Aged	3	0	5	−14	0.38	0.15	860	530	330 (−, 470)	61 (45, 110)	3.8
91	1	50°02'48"	124°22'41"	Fresh	8	0	5	−25	0.23	0.08	250	200	160 (110, 200)	35 (18, 55)	2.1
	2			Aged		5	10	−25	0.18	0.15	−	−	−	−	−
92	1	50°39'07"	122°23'53"	Fresh	14	0	5	−16	0.17	0.08	80	70	50 (30, 70)	34 (15, 60)	2.3
	2			Aged		5	10	−16	0.18	1.25	110	100	80 (50, 100)	29 (11, 55)	2.2
93	1	50°24'50"	124°54'20"	Fresh	8	0	1	−	−	0.07	170	100	70 (40, 90)	53 (39, 73)	2.5
	2			Aged		2	6	−21	0.21	0.175	310	140	100 (50, 130)	68 (59, 85)	2.7
94	1	50°09'05"	125°46'06"	Fresh	8	0	1	−24	−	0.1	1200	400	260 (110, 360)	78 (69, 91)	2.8
95	1	50°54'43"	127°04'50"	Aged	18	0	3	−22	0.23	0.175	280	110	80 (40, 100)	73 (64, 86)	2.6
	2			Aged		3	8	−23	0.18	0.9	230	80	50 (30, 70)	77 (68, 89)	2.7
	3			Aged		8	13	−25	0.27	1	90	50	30 (10, 40)	71 (55, 92)	3.2
	4			Aged		13	18	−25	0.14	1.3	520	310	130 (−, 240)	75 (54, 148)	4.8
96	1	49°14'21"	129°43'13"	Fresh	13	0	5	−22	0.37	0.08	1300	390	240 (20, 330)	83 (76, 99)	3.4
	2			Aged		5	10	−23	0.25	0.6	230	90	50 (10, 70)	81 (69, 98)	3.3
	3			Aged		10	15	−22	0.22	1	370	130	80 (30, 110)	80 (71, 93)	3.0
97	1	47°39'18"	131°13'10"	Aged	20	0	5	−22	0.36	0.2	1000	360	240 (90, 310)	76 (69, 91)	2.8
	2			Aged		5	10	−18	0.27	1.1	160	80	50 (20, 70)	71 (57, 91)	3.0

Site	Layer	Latitude N	Longitude E	Snow type	Site average snow depth (cm)	Sample depth (cm) Top	Bottom	Temperature (°C)	Snow density (g cm⁻³)	R_m (mm)	C_{BC}^{equiv} (ng g⁻¹)	C_{BC}^{max} (ng g⁻¹)	C_{BC}^{est} (ng g⁻¹)	f_{non-BC}^{est} (%)	$\mathring{A}_{tot}$ 450:600 nm
	3			Aged		10	15	-14	0.32	0.9	30	20	10 (5, 20)	74 (44, 90)	3.1
98	1	45°25'38"	130°58'55"	Fresh	35	0	5	-20	0.22	0.09	2200	2100	1500 (750, 1800)	32 (16, 65)	2.5
	2			Aged		5	10	-23	0.24	0.5	640	350	240 (120, 310)	58 (46, 79)	2.6
	3			Aged		10	15	-15	0.24	0.75		60	20 (10, 50)	74 (52, 90)	3.1
	4			Aged		15	20	-14	0.26	0.75	60	40	20 (10, 30)	75 (48, 89)	3.1
	5			Aged		20	25	-13	0.28	1.1	430	260	170 (60, 220)	62 (49, 86)	2.8
	6			Aged		25	30	-11	0.29	1.3	350	170	110 (70, 150)	68 (58, 81)	2.5
	7			Aged		30	35	–	0.33	1.3	290	120	70 (40, 100)	75 (65, 87)	2.6
99	1	43°36'10"	125°42'04"	Fresh	13	0	5	-2	0.18	0.085	1500	920	640 (240, 820)	54 (40, 84)	2.8
	2			Aged		5	12	-3	0.23	1.1	5800	3800	2900 (1500, 3600)	51 (39, 74)	2.5
	3			Aged		12	18	-4	0.28	1.1	2800	1400	1100 (630, 1300)	62 (53, 77)	2.3
	4			Aged		18	24	-4	0.28	0.7	2000	1500	1100 (540, 1400)	46 (32, 73)	2.6
100	1	43°30'45"	127°15'34"	Fresh	18	0	2.5	–	0.14	0.075	1700	700	510 (250, 650)	71 (63, 86)	2.6
	2			Aged		2.5	4	-2	0.18	0.25	4100	2900	1700 (–, 2400)	58 (43, 103)	3.6
	3			Aged		4	9	-2	0.23	0.9	1100	560	410 (210, 530)	60 (49, 80)	2.6
	4			Aged		9	15	-3	0.24	0.8	1600	730	540 (270, 690)	66 (57, 83)	2.6
101	1	43°47'25"	125°46'08"	Fresh	22	0	5	-19	0.24	0.07	5600	5100	3700 (1300, 4600)	35 (18, 76)	2.9
	2			Aged		5	10	-17	0.24	0.9	4300	3800	2600 (880, 3300)	40 (24, 79)	2.9
	3			Aged		10	15	-15	0.23	1.1	660	390	310 (210, 390)	46 (33, 63)	2.1
	4			Aged		15	20	-14	0.28	1.3	300	250	190 (110, 240)	39 (21, 67)	2.5
102	1	42°12'34"	126°37'50"	Fresh	46	0	3	-8	0.13	0.07	2100	1700	1400 (810, 1800)	37 (19, 61)	2.3
	2			Aged		3	8	-8	0.26	0.1	590	380	280 (150, 370)	53 (37, 75)	2.6
	3			Aged		8	13	-8	0.24	0.3	2500	2100	1600 (430, 2100)	35 (15, 81)	3.1

Site	Layer	Latitude N	Longitude E	Snow type	Site average snow depth (cm)	Sample depth (cm) Top	Bottom	Temperature (°C)	Snow density (g cm⁻³)	R_m (mm)	C_{BC}^{equiv} (ng g⁻¹)	C_{BC}^{max} (ng g⁻¹)	C_{BC}^{est} (ng g⁻¹)	f_{non-BC}^{est} (%)	Å_{tot} 450:600 nm
	4			Aged		13	18	-8	0.28	0.6	1900	1100	770 (230, 1000)	60 (48, 88)	3.0
	5			Aged		18	23	-7	0.32	1	1100	630	450 (200, 590)	60 (47, 82)	2.7
	6			Aged		23	28	-6	0.27	1.2	1500	1300	900 (300, 1200)	38 (21, 80)	2.9
	7			Aged		28	33	-5	0.26	1.2	860	490	340 (130, 460)	66 (53, 87)	2.9
	8			Aged		33	38	-5	0.3	1	430	200	110 (30, 160)	74 (61, 95)	3.2
	9			Aged		38	43	-4	0.29	0.8	520	250	160 (70, 220)	71 (58, 87)	2.8

Table 2. Chemical species (ng g^{-1}) in surface snow for sites across northeastern China in January 2014. The datasets of SO_4^{2-}, NO_3^-, and NH_4^+ were reprinted from Wang et al. (2015).

Site	MD	BC	OC	$K^+_{biosmoke}$	SO_4^{2-}	NO_3^-	NH_4^+	Sea salt
90	1900	380	6700	327	1685	213	22	868
91	1700	180	590	179	853	465	36	827
92	1300	60	280	150	511	105	19	456
93	1700	80	450	213	718	387	90	960
94	3300	300	2700	118	1335	550	28	554
95	2000	90	600	164	587	523	39	669
96	2300	280	3900	309	1285	493	91	1227
97	2400	280	2400	173	1163	407	38	753
98	3900	1600	13300	633	3096	747	195	2516
99	3000	770	4700	372	3379	1492	155	2310
100	3800	570	4000	260	4237	2258	487	2195
101	3500	4200	32000	1337	12382	2364	–	5131
102	5800	1700	2400	488	8034	3631	769	4420
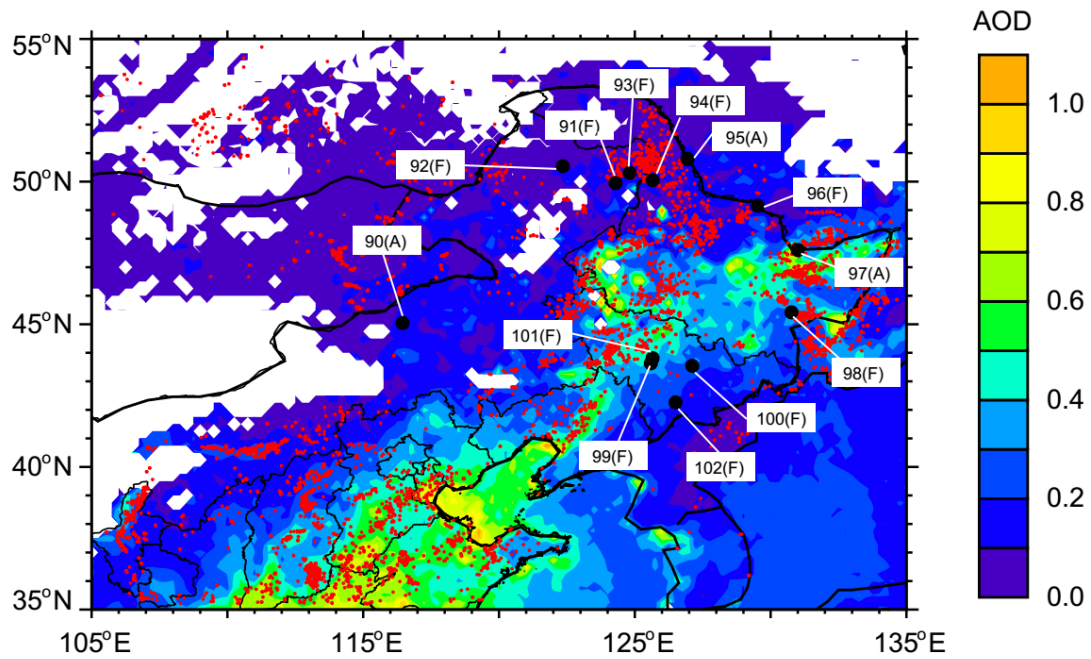
Fig. 1. Spatial distribution of the averaged AOD retrieved from Aqua-MODIS over northern China from October 2013 to January 2014. The red dots are MODIS active fire locations, the black dots are the sampling locations. The site numbers beginning at 90 in this study are numbered in chronological order followed by Wang et al. (2013a), and Ye et al. (2012). The "A" and "F" refer to aged snow and fresh snow, respectively.
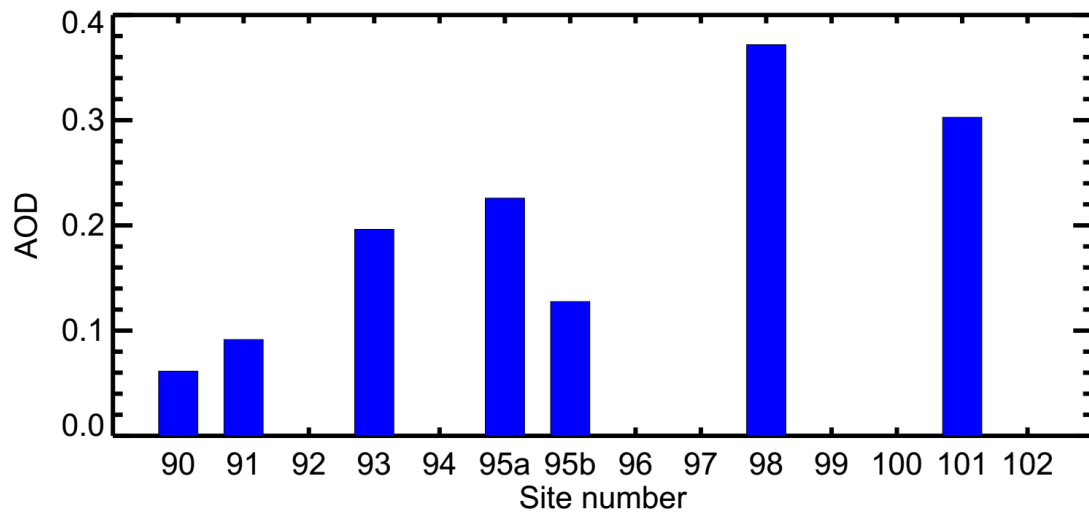
Fig. 2. The variation in AOD at 500 nm at different sites measured using a Microtops Ⅱ Sun photometer over northeastern China in January 2014.
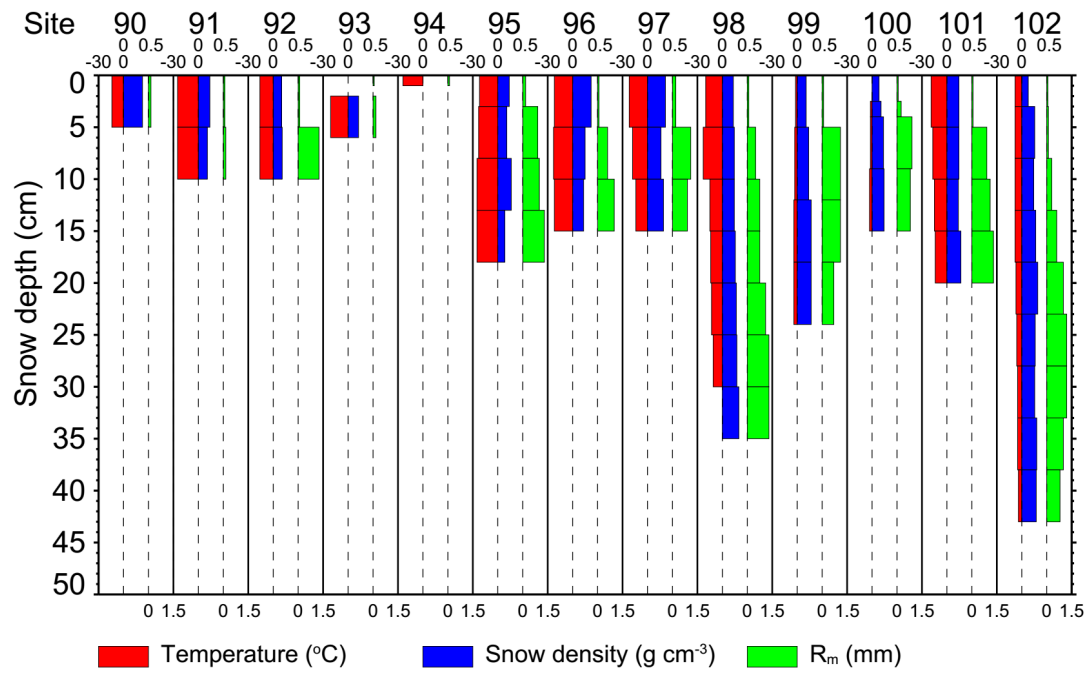
Fig. 3. Vertical temperature, snow density, and measured snow grain radius (R_m) profiles at each site during the 2014 Chinese snow survey.
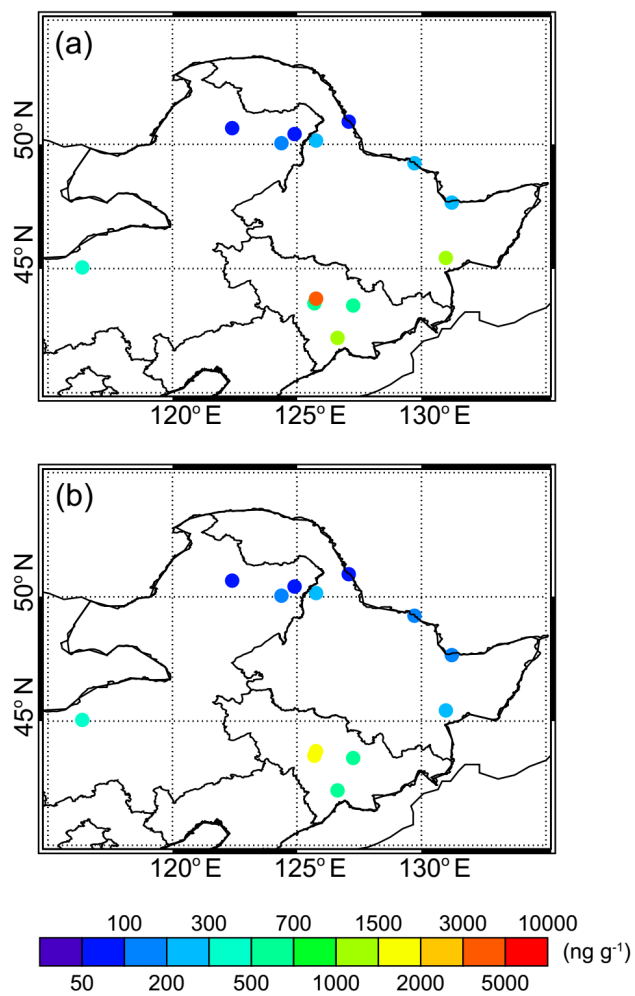
Fig. 4. The spatial distribution of C_{BC}^{est} in the (a) surface and (b) average snow in 2014 across northeastern China.
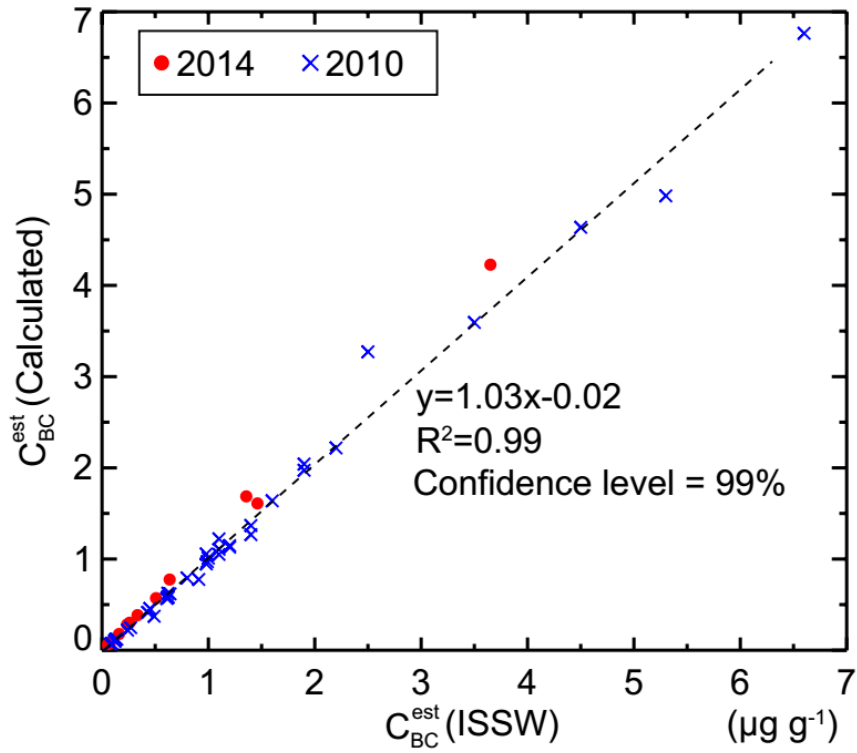
Fig. 5. Comparisons between the calculated and optically measured C_{BC}^{est} in surface snow during 2010 and 2014 snow surveys. The datasets of measured C_{BC}^{est} in 2010 from sites 3-40 were reprinted from Wang et al. (2013a).
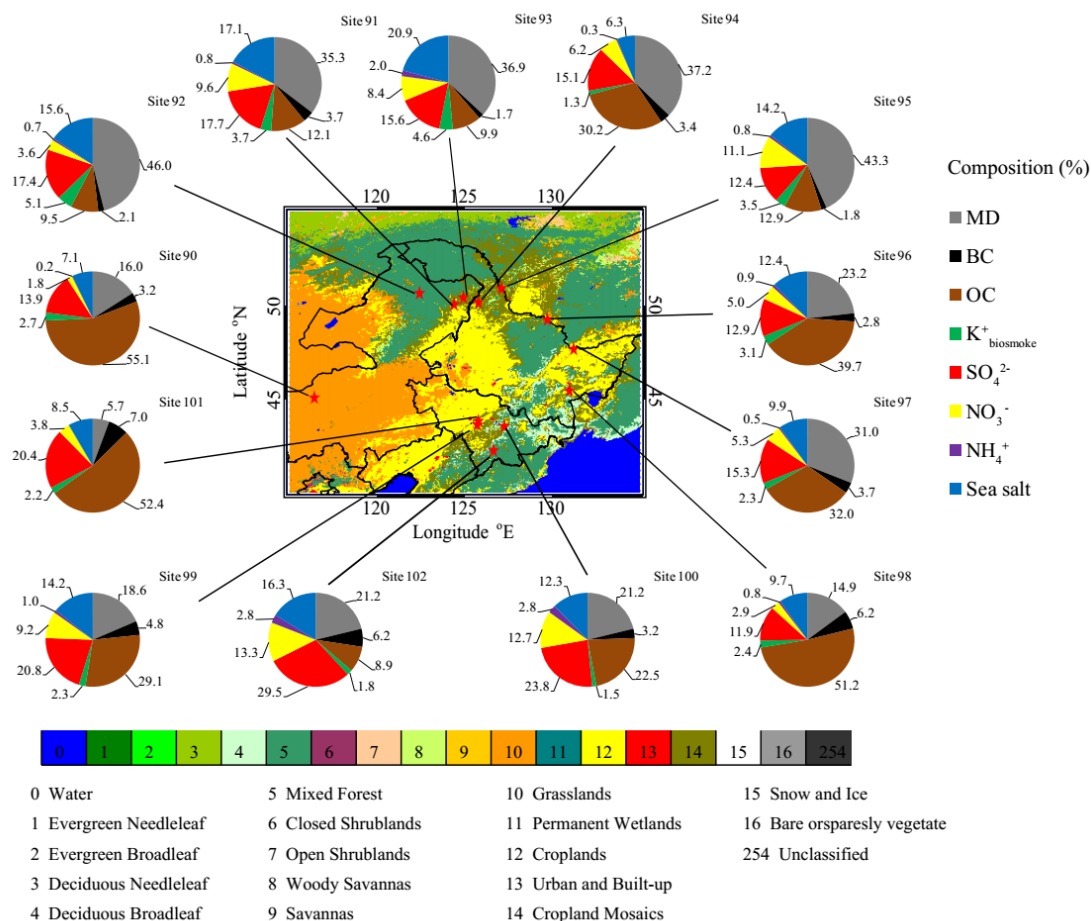
Fig. 6. The major components include MD, BC, OC, $K^+_{Biosmoke}$, secondary ions (SO_4^{2-}, NO_3^-, and NH_4^+), and sea salt in the surface snow samples collected in January 2014. The distribution of 17 different surface vegetation types retrieved from MODIS global land cover type product (MCD12C1) with 0.05 spatial resolution was used in this study. The datasets of SO_4^{2-}, NO_3^-, and NH_4^+ were reprinted from Wang et al. (2015).
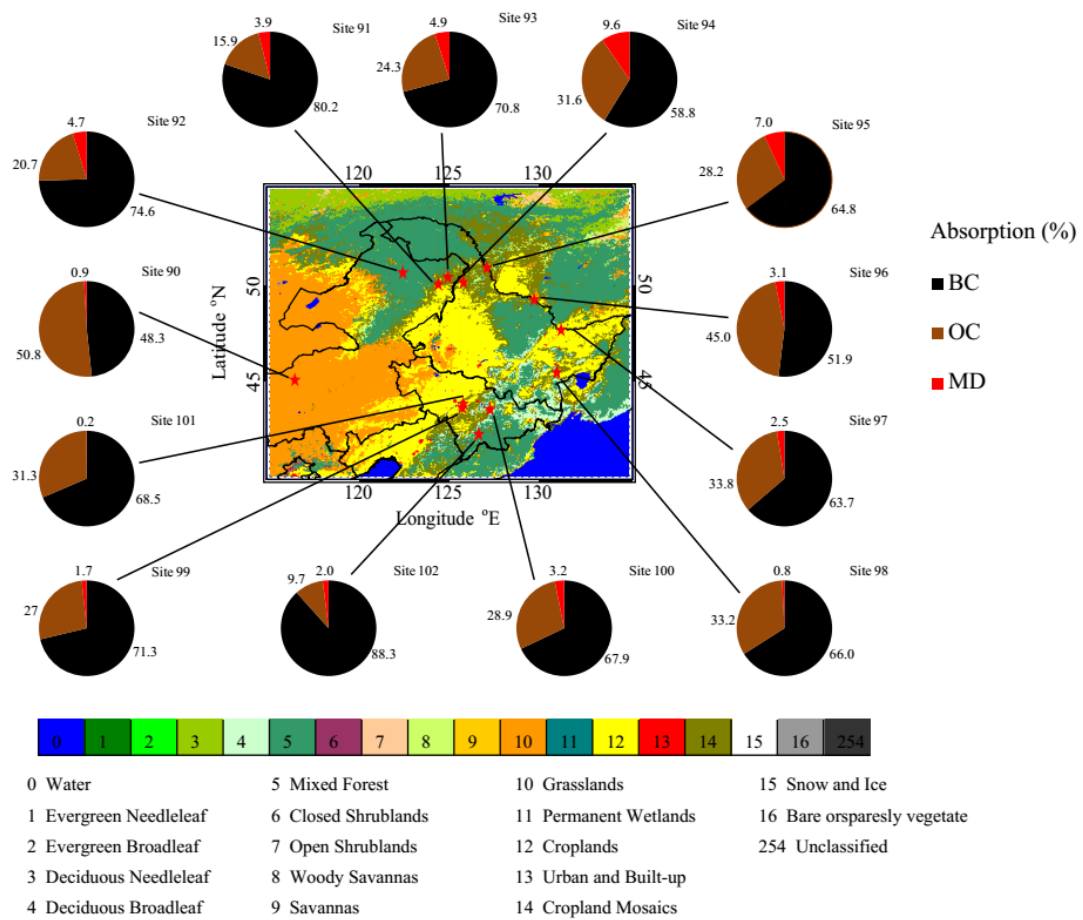
Fig. 7. The light absorption of ILAPs in surface snow in January 2014. The distribution of 17 different surface vegetation types retrieved from MODIS global land cover type product (MCD12C1) with 0.05 spatial resolution was used in this study.
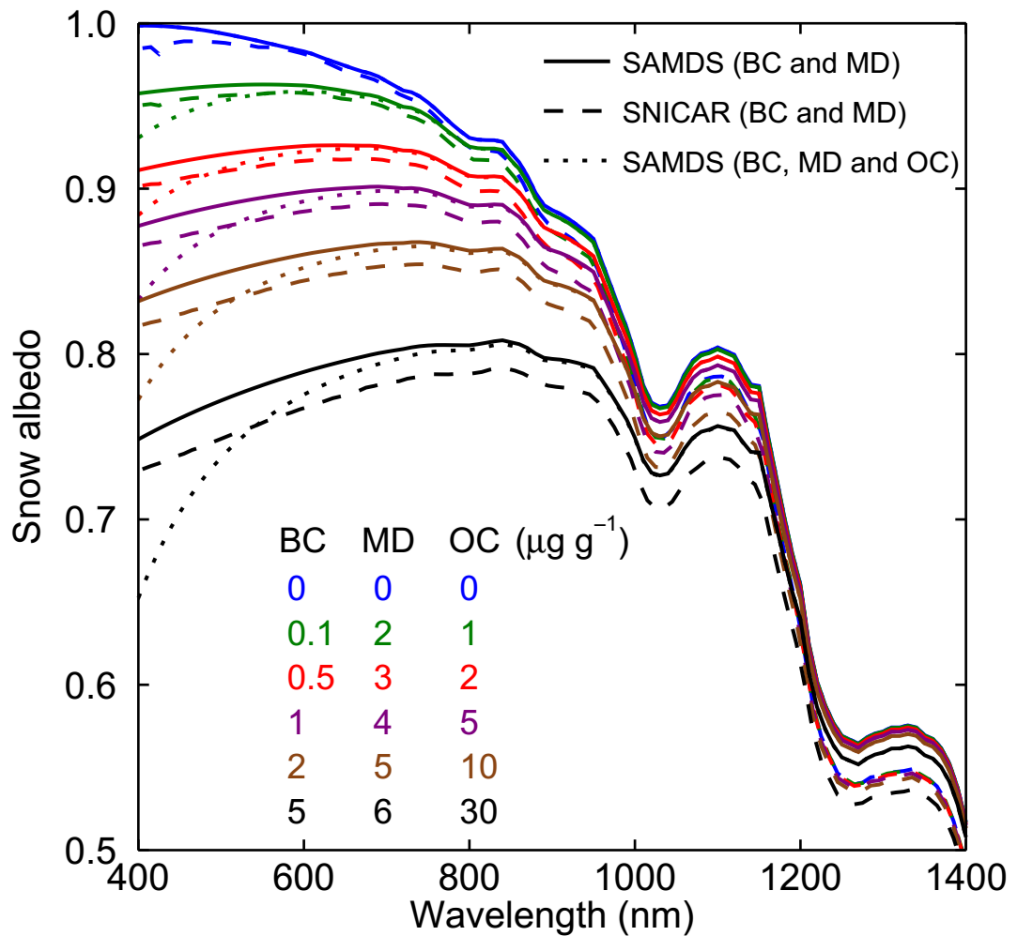
Fig. 8. Spectral albedo of snow with different contaminants for a 60° solar zenith angle and a 100 μm R_{eff}. Solid and dashed lines show the SAMDS and SNICAR model predictions for BC and MD. Dotted lines show the SAMDS model predictions for all ILAPs, including BC, MD, and OC.
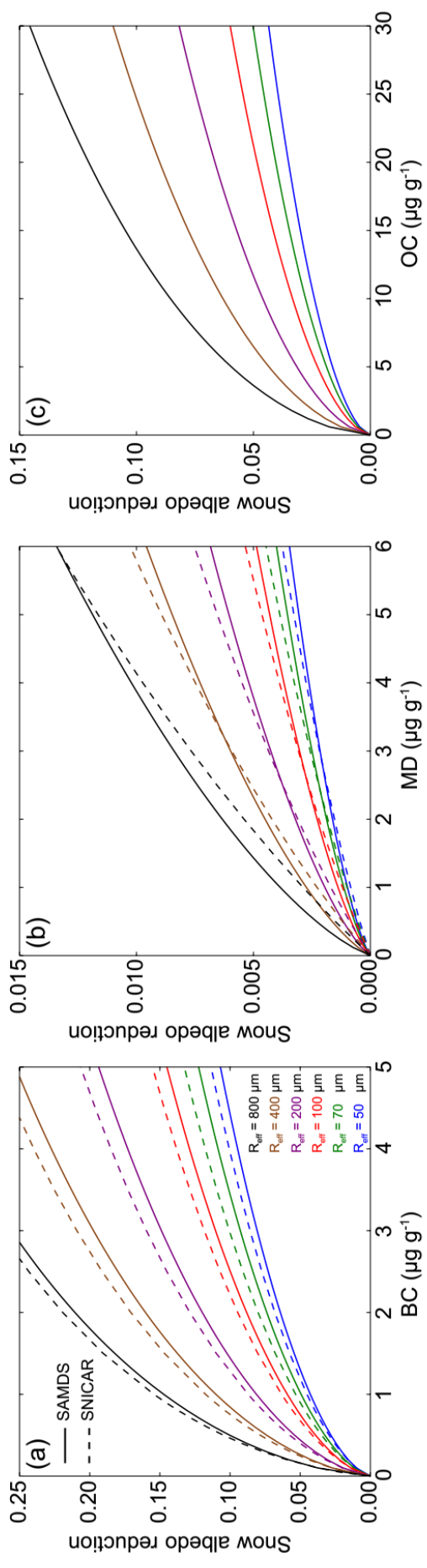
Fig. 9. Spectrally weighted snow albedo reduction over the 400–1400 nm solar spectrum attributed to (a) BC, (b) MD, and (c) OC computed as the albedo of pure snow minus the albedo of contaminated snow for a 60° solar zenith angle. Solid and dashed lines show the SAMDS and SNICAR models predictions. The MAC values of BC, OC, and Fe were assumed to be 7.5 m^2 g^{-1}, 0.3 m^2 g^{-1}, and 0.9 m^2 g^{-1} at 550 nm, respectively, in SAMDS model.
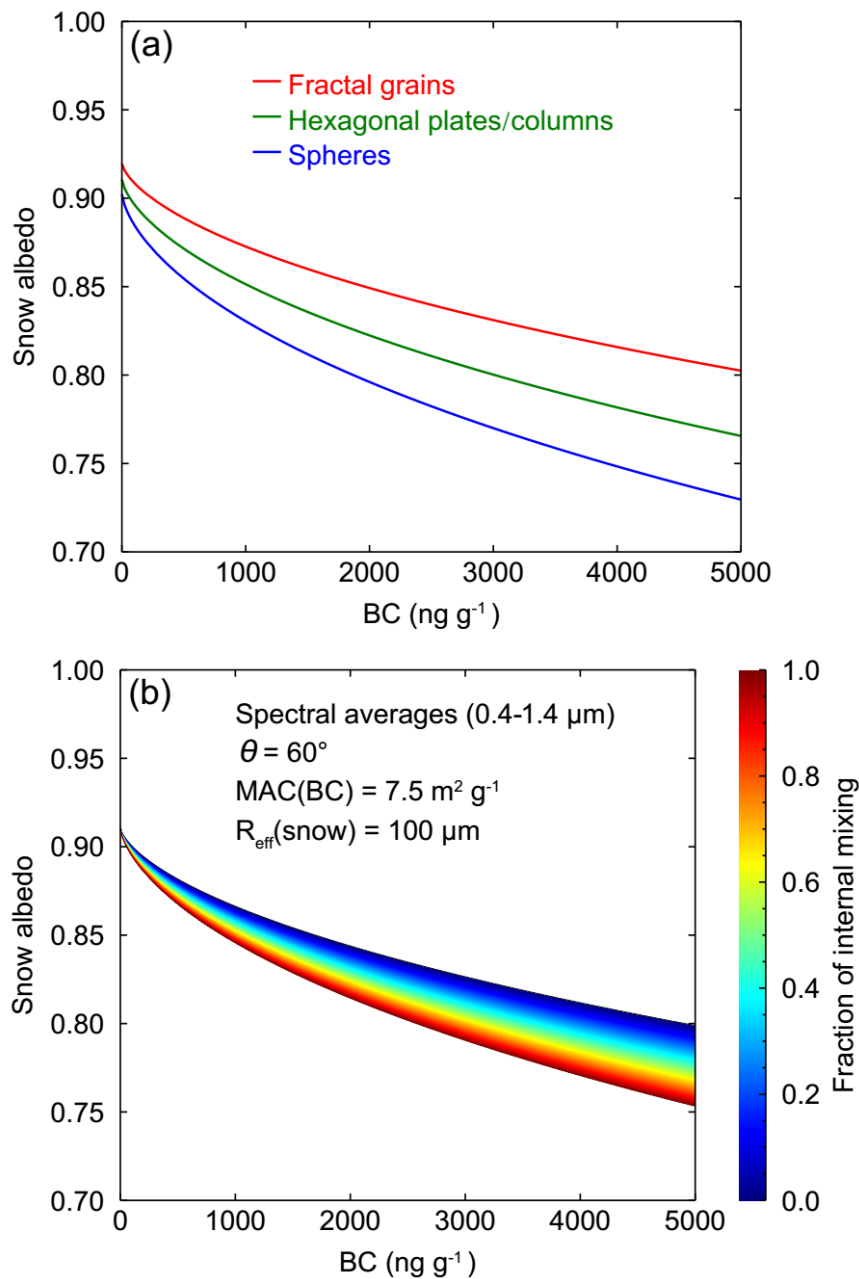
Fig. 10. Spectral albedo of snow as a function of BC mixing ratios in snow by using SMDAS model for: (a) the irregular morphology of snow grains (fractal grains, hexagonal plates/columns, and spheres), (b) internal/external mixing of BC and hexagonal plates/columns snow grains. Also shown are model parameters including integrate spectral wavelengths (400-1400 nm), solar zenith angle (θ), mass absorption coefficient (MAC) of BC, and snow grain optical effective radius (R_{eff}).
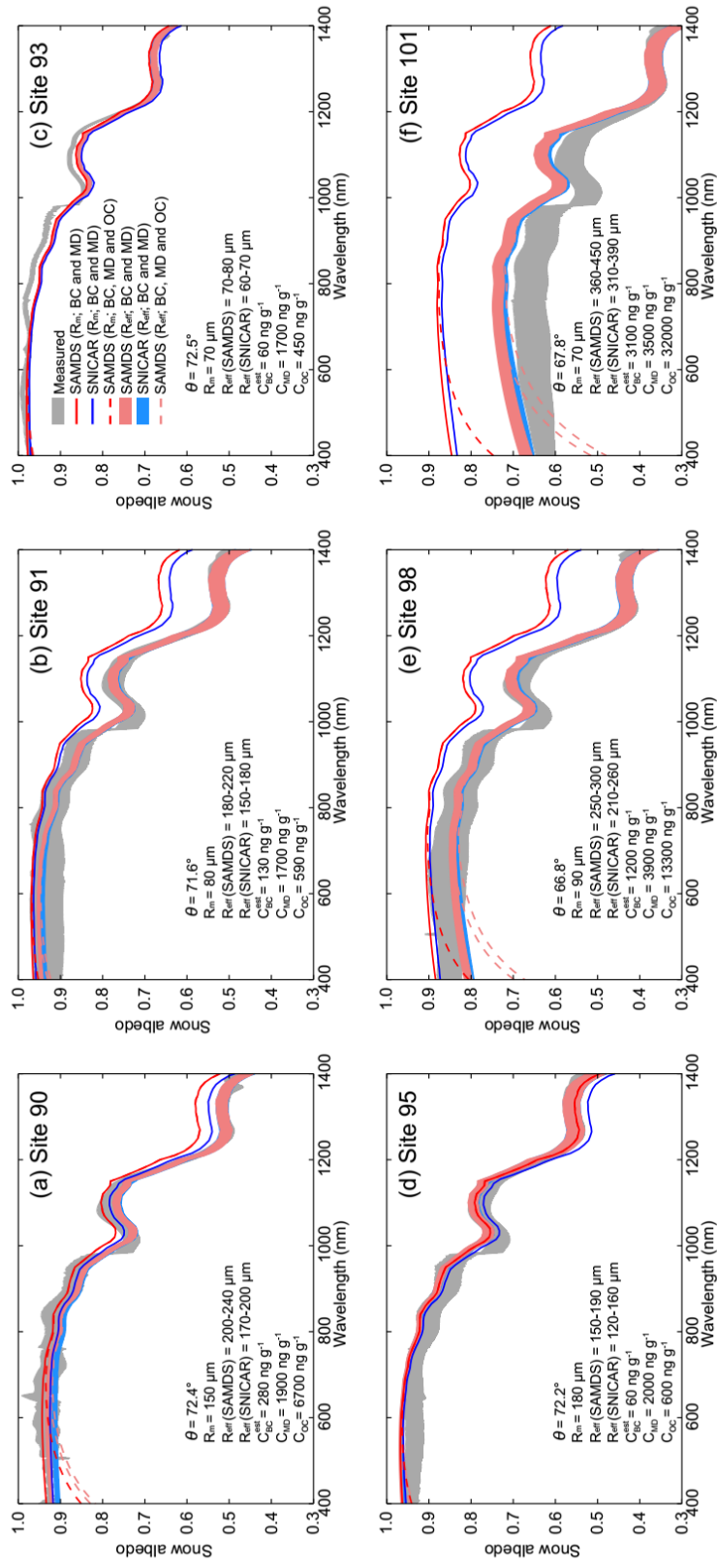
Fig. 11. Measured and modeled spectral albedos of snow at sites (a) 90, (b) 91, (c) 93, (d) 95, (e) 98, and (f) 101. Gray shaded bands correspond to measured spectral albedos using an spectroradiometer. Red and blue solid lines correspond to spectral albedos simulated by the SAMDS and SNICAR models with measured snow grain radii (R_m), and light red and blue shaded bands correspond to the albedos with calculated snow grain optical effective radii (R_{eff}). Contaminants only include BC and MD in the SAMDS and SNICAR models. In the SNICAR model, the ratio of Fe in dust was assumed to be 2.8%. Dashed red lines are similar to solid red lines, although OC should be added to the list of contaminants in the SAMDS model.

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