Fluorescence characteristics, absorption properties, and radiative effects of water-soluble organic carbon in seasonal snow across northeastern China

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Abstract. ~~Although w~~Water-soluble organic carbon (WSOC) in the cryosphere can significantly influence the global carbon cycle and radiation budget~~.,~~ However, WSOC in the snowpack has received little scientific attention to date. This study reports the fluorescence characteristics, absorption properties, and radiative effects of WSOC based on 34 snow samples collected from sites in northeastern China. ~~Sampling sites were divided into five groups, comprising southeastern Inner Mongolia (SEIM), northeastern Inner Mongolia (NEIM), the south of northeastern China (SNC), the north of northeastern China (NNC), and the Changbai Mountain area (CBM).~~ We found ~~Together, these groups represent~~ a significant degree of regional WSOC variability, with concentrations ranging from ~~0.50~~0.5 ± ~~0.19~~0.2 to ~~5.70~~5.7 ± ~~3.68~~3.7 µg g^{-1} (mean = ~~3.59~~3.6 ± ~~3.19~~3.2 µg g^{-1}). ~~We t~~Then he three principal fluorescent components of WSOC are identified as~~the three principal fluorescent components: of WSOC as~~ (1) the~~a~~ high-oxygenated ~~humic-like component~~ humic-like substances (HULIS-1) of terrestrial origin, (2) the~~a~~ low-oxygenated humic-like ~~component~~ substances (HULIS-2) of mixed origin, and (3) ~~and the~~a protein-like ~~component~~substances (PRLIS) derived from autochthonous microbial activity. In southeastern Inner Mongolia (SEIM)~~SEIM~~, a region dominated by desert and exposed soils, the WSOC ~~content~~ exhibits the highest humification index (HIX) but the lowest fluorescence (FI) and biological (BIX) indices; the fluorescence signal is mainly attributed to HULIS-1, and thus implicates soil as the primary source. By

contrast, the HIX (FI and BIX) value ~~was~~ is the lowest (highest) and percentage of PRLIS is the ~~most~~

~~intense~~highest in the remote ~~grasslands and forested~~ area~~s~~ of the northeastern Inner Mongolia

(NEIM)~~NEIM~~, suggesting a primarily biological source. For south and north of northeastern China (SNC

and NNC)~~SNC and NNC~~, both of which are characterized by intensive agriculture and industrial activity,

the fluorescence signal is dominated by HULIS-2, and the HIX, FI, and BIX values are all moderate,

indicating the mixed origins for WSOC (anthropogenic activity, microbial activity, and soil). We also

observed that, throughout northeastern China, the light absorption of WSOC is dominated by HULIS-1,

followed by HULIS-2 and PRLIS. The contribution of WSOC to albedo reduction (average concentration

3.6 μg g^{-1}) in the ultraviolet–visible ~~(UV–vis)~~ band is approximately half that of black carbon (BC:

average concentration 0.6 μg g^{-1})~~.;~~ ~~R~~radiative forcing is 3.8 (0.8) W m^{-2} in old (fresh) snow, equating to

19 % (17 %) of the radiative forcing of BC. These results indicate that WSOC has a profound impact on

snow albedo and the solar radiation balance.

1   Introduction

Seasonal snow plays a significant role in Earth's solar radiation energy budget owing to its high

reflectivity (Beniston et al., 2017; Usha et al., 2020; Xie et al., 2018). In recent decades, however, the

extent of snow-covered areas has trended downward, partially as a result of the presence of light-

absorbing particles (LAPs) in the snowpack (Barnett et al., 2008; Dumont et al., 2014; Groisman et al.,

1994~~; Dumont et al., 2014~~). ~~Black carbon (BC), organic carbon (OC), mineral dust (MD), and biota~~

~~comprise the principal light absorbing particles in seasonal snow (Di Mauro, 2020; Qian et al., 2015),~~

~~which together serve to lower surface albedo and impose a positive radiative forcing (Dumont et al.,~~

~~2014; Hansen and Nazarenko, 2004; Warren and Wiscombe, 1980; Zhang et al., 2017). Concurrently,~~

~~LAPs absorb solar radiation, thereby accelerating snow melt (Li et al., 2021b). Ultimately, the disruption~~

of the global radiative balance due to LAPs has important implications for regional and global climate change (Skiles et al., 2018). The LAPs in seasonal snow, such as black carbon (BC), organic carbon (OC), mineral dust (MD), and biota (Beres et al., 2020; Di Mauro, 2020; Els et al., 2020; Qian et al., 2015; Wu et al., 2016), can strongly absorb solar radiation, which together serves to lower surface albedo and impose a positive radiative forcing (Cui et al., 2021; Dumont et al., 2014; Hansen and Nazarenko, 2004; Shi et al., 2022b; Warren and Wiscombe, 1980; Zhang et al., 2017). Ultimately, LAPs can accelerate snow melting (Li et al., 2021b) and disturb the global radiative balance, therefore, have important implications for regional and global climate change (Shi et al., 2022a; Skiles et al., 2018; Shi et al., 2022a).

Snowpack BC and MD have been the focus of considerable research in snow-covered regions worldwide (Antony et al., 2014; Doherty et al., 2014; Hegg et al., 2010; Li et al., 2021a; Wang et al., 2014b; Zhang et al., 2018; Antony et al., 2014; Hegg et al., 2010; Doherty et al., 2014; Wang et al., 2014b). As the most important LAP (Bond et al., 2013; Doherty et al., 2010; Shi et al., 2020; Wang et al., 2014b; Shi et al., 2020), the radiative efficiency of snowpack BC can be more than three times greater than that of carbon dioxide (Flanner et al., 2007). , and MD, another important snowpack LAP, is also known to alter the cryospheric environment owing to its light-absorbing properties (Di Mauro et al., 2015; Painter et al., 2007; Sarangi et al., 2020; Shi et al., 2021). Recently, researchers have also begun evaluating the influence of biomes on global snow albedo (Hotaling et al., 2021). In contrast, however, the role of OC remains poorly understood because of its complex composition and a relative dearthlack of OC-focused research. Consequently, substantial uncertainty surrounds the origins, optical properties, and radiative effects of snowpack OC.

1 ~~A recent study has reported that the storage of OC in mountain glaciers and ice caps (~11 % of Earth's~~

2 ~~land surface) could be as high as 6 petagrams (Pg; Hood et al., 2015), the majority of which is water-~~

3 ~~soluble organic carbon (WSOC). WSOC is one of the largest sources of bioavailable organic carbon in~~

4 ~~aquatic ecosystems (Battin et al., 2009). Moreover,~~ ~~The recent study has~~A recent study has reported that

5 the storage of OC in mountain glaciers and ice caps (~11 % of Earth's land surface) could be as high as

6 6 petagrams (Pg, Hood et al., 2015), the majority of which is water-soluble organic carbon (WSOC)

7 (~~Hood et al., 2015;~~ Yan et al., 2016). The substantial part of WSOC in the glacier is highly bioavailable

8 and can be a source of labile organic matter for downstream ecosystems (Singer et al., 2012). The

9 physical and photochemical processes can occur within various WSOC in snow cover and glaciers, and

10 therefore have a great effect on atmospheric and glacier chemistry (Antony et al., 2011; Domine, 2002;

11 Grannas et al., 2007~~; Antony et al., 2011~~). Moreover, WSOC has important influences on the energy

12 budget and radiative forcing of snow cover and glaciers (Kirillova et al., 2014; Ram et al., 2010; Yan et

13 al., 2016). As~~as~~ the chief absorber of WSOC, water-soluble brown carbon (WS-BrC) can absorb

14 significant measures of solar radiation in the ultraviolet–visible (UV–vis) wavelengths (Murphy et al.,

15 2008). For instance, in their analysis of 21 snow samples from the Arctic and Antarctic ~~snow samples~~,

16 Anastasio and Robles (2007) ~~observed that~~ attributed 50 % of the total light absorption coefficients at

17 wavelengths of > 280 nm ~~might be attributed~~ to organic chromophores of WSOC. ~~In surficial snow~~

18 ~~samples from Barrow, Alaska,~~ Beine et al. (2011) reported that WSOC occupies almost the entire

19 absorption spectrum of the photochemically active region (300–450 nm) in surficial snow samples from

20 Barrow, Alaska.~~, and~~ ~~And~~ Feng et al. (2016) observed that absorption in cryoconite samples from the

21 central Tibetan Plateau is dominated by WSOC components in the 300–350 nm range. Similarly, Yan et

22 al. (2016) measured WSOC in glacial snow from Laohugou glacier, northern Tibetan Plateau, where they

found that the radiative forcing is ~10 % that of BC. Together, these studies indicate that WSOC plays a

key role in global snowpack energy absorption (Niu et al., 2018; Zhang et al., 2020). We note that recent

researches on cryospheric WSOC mainly focused on alpine glaciers and polar regions (Guo et al., 2022;

Li et al., 2022; Guo et al., 2022), while the extensive mid-latitude regions impacted by seasonal snowpack

remain relatively understudied.

The composition of WSOC is typically complex, and the characteristics of fluorescence and absorption

can vary widely among the different components. Nonetheless, recent studies have tended to treat WSOC

as a single entity and focus on the overall impacts (Barrett and Sheesley, 2017; D'Sa et al., 2014; Niu et

al., 2018; Wu et al., 2019), such that the specific roles of individual components are poorly constrained.

One commonly used analytical method for distinguishing the components and properties of fluorescence

is the fluorescence excitation-emission matrixees (EEMs), which has the advantage of high sensitivity

and small sample size (Coble, 1996; Kowalczuk et al., 2005). First applied in oceanic contexts (Coble et

al., 1990), EEMs has have been gradually extended to lakes, fog water, and rainwater, and atmospheric

aerosols in addition to glacial meltwater, ice cores, and snow (Birdwell and Valsaraj, 2010; Huguet et

al., 2009; McKnight et al., 2001). At present, the application of EEMs on atmospheric aerosols has

entered a mature stage. EEMs have been used to identify fluorescent WSOC components in aerosols

from polar regions or urban backgrounds, and it has been found that different structures of WSOC

fractions exhibit different oxidation properties, which may provide a clue to understand the chemical

formation or loss of organic chromophores in atmospheric aerosols (Chen et al., 2016; Fu et al., 2015).

Recently, this method has been gradually extended to the analysis of glacier samples and snow samples

(Feng et al., 2018; Guo et al., 2022; Zhou et al., 2019b). Concurrently, parallel factor analysis

(PARAFAC) is an effective approach for extracting from complex EEMs the individual fluorescence

components and their corresponding fluorescence information, thus making EEM–PARAFAC a direct and viable means for exploring sources of WSOC. For example, Zhou et al., (2019b) used EEM–PARAFAC to identify the multiple sources of WSOC measured in seasonal snow in northwestern China. Accordingly, we have applied EEM–PARAFAC in our analysis of snow samples for this study.

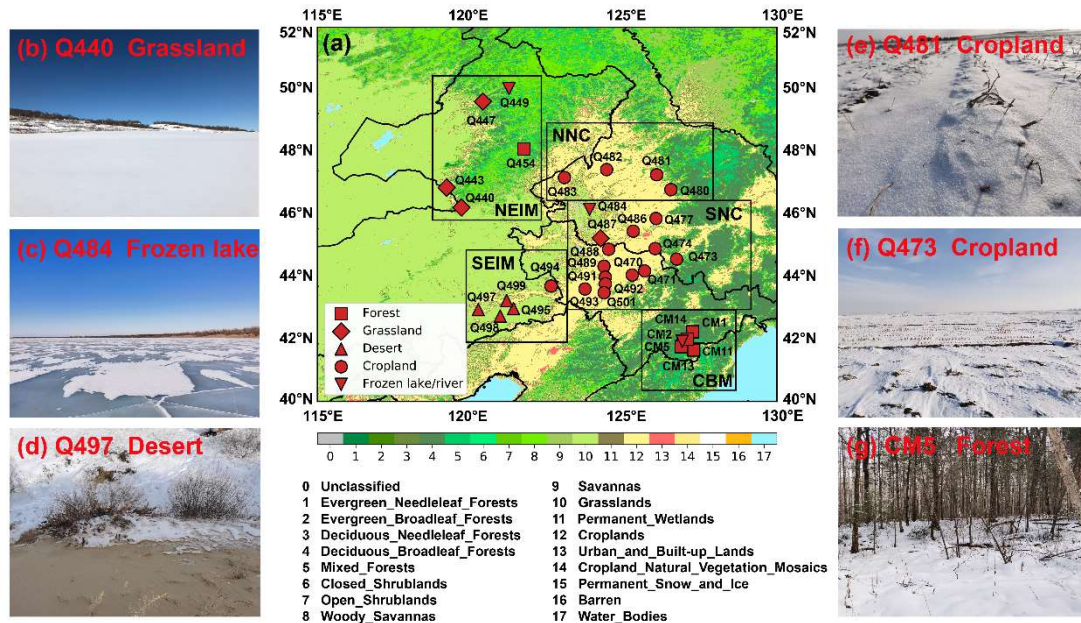
EEM-PARAFAC can only provide plausible information about the component-specific influence of WSOC on fluorescent properties, and the quantitative fractional contributions of specific components to light absorption are still unknown. Recently, Chen et al. (2019a) collected atmospheric aerosol samples in $PM_{2.5}$ over Xi'an, China, and successfully attributed the dithiothreitol (DTT) activity levels to various BrC components by coupling DDT and BrC datasets. A similar attribution method has been applied to various research areas, including climate change, extreme weather, and atmospheric environments (Cao et al., 2015; Pokrovsky, 2019; Xin et al., 2016; Zhao et al., 2019). In this study, we applied a multiple linear regression method comparable to that of Chen et al. (2019a) to derive the fractional contribution of each WSOC component to light absorption. Despite this method having been used elsewhere (Wu et al., 2022; Wu et al., 2021), it remains a highly innovative approach to evaluating the light absorption of snowpack WSOC.

Northeastern China supports an extensive snowpack during winter and spring. As a major industrial and agricultural center, this region is also the principal source of heavy airborne pollutants that are incorporated into seasonal snow via wet and dry deposition (Wang et al., 2017). Coupled with intensive tilling of farmland, the geographical proximity of northeastern China to the neighboring desert regions also provides a source of soil organic matter that becomes entrained into the snowpack (Wang et al., 2013b). Compared with research on BC-snow mixing ratios and their radiative impact in northeastern China (Dang et al., 2017; Huang et al., 2011; Pu et al., 2019), studies of WSOC are still in their infancy.

To address this deficiency, we ~~analyzed 34 samples of seasonal snow collected in December 2020 and~~ ~~January 2021 to~~ ~~made~~make the ~~first~~~~primary~~first investigation of the fluorescence characteristics, absorption properties, and radiative effects of WSOC in seasonal snow samples in northeastern China. Specifically, we applied EEM–PARAFAC to identify the origins and fluorescence characteristics of snowpack WSOC, after which we derived individual absorption contributions for each WSOC component using fluorescence data, an absorption data series, and an attribution method. Finally, we estimated the reduction of snow albedo and radiative forcing caused by WSOC and BC via the Spectral Albedo Model for Dirty Snow (SAMDS) radiative transfer model.

2   Methods

2.1   Sample collection

During ~~the months of~~January and December 2020 and January 2021, we collected 34 snow samples from sites across northeastern China, including the eastern part of Inner Mongolia and Heilongjiang and Jilin provinces. Sample numbers were set following previous campaigns (Pu et al., 2017; Wang et al., 2013b, 2017), with the exception that samples from the Changbai Mountain area are numbered individually. The geographical distribution of sampling sites and respective land-cover types are shown in Figure 1a.~~;~~ ~~our~~The sites are characterized by five land-cover types, including forest, grassland, desert, cropland, and frozen lake/river (Fig. 1b–g). On the basis of these geographical and environmental classifications, we divided the sampling sites into five broad regions: southeastern Inner Mongolia (SEIM; Q494–495, Q497–499), the south of northeast China (SNC; Q470–471, Q473–474, Q477, Q484, Q486–Q489, Q491–Q493, Q501), the north of ~~~~northeast China (NNC; Q480–483), the Changbai Mountain area (CBM; CM1–CM2, CM5, CM11, CM13–CM14), and northeastern Inner Mongolia (NEIM; Q440, Q443, Q447, Q449, Q454).

2   **Figure1: (a) Information on sampling site distributions in northeastern China, including the land cover type,**

3   **site number, and grouping. Land cover types are derived from Collection 5.1 of the MODIS global land cover**

4   **type dataset (MCD12Q1: https://lpdaac.usgs.gov/products/mcd12q1v006/) and are indicated by specific**

5   **colors and symbols relative to sampling sites. Sampling sites are divided into ~~the~~ five groups defined by black**

6   **rectangles. (b–g) Photographs depicting the typical snow and ground-cover conditions of our various sampling**

7   **sites.**

8   ~~Our~~The ~~sample~~ sampling sites were chosen at random but had to be located ≥ 20 km from cities and

9   villages and at least 1~~a~~ km~~kilometer~~ upwind of roads or railroads to minimize the influence of single-

10   point pollution sources and to ensure the broadest regional representation. Furthermore, we performed

11   sample collection oriented toward the wind to avoid contamination from personnel. At each site, we used

12   a sterile disposable shovel to collect 0–5 cm-thick samples of surface snow, which were subsequently

13   stored in sterile Whirlpak (Nasco, WI, USA) bags. For snow depths of < 5 cm, we determined the

14   sampling depth according to the actual conditions to avoid inducting significant soil impurities during

15   sampling. Snow samples were melted at room temperature (25 °C) and stored in a freezer at −20 °C until

16   analysis in the laboratory. For more operational details, we refer the reader to Wang et al. (2013b).

2.2 Chemical species analysis

~~After collection, samples were melted at room temperature (25 °C) before being filtered using a~~ ~~disposable sterile syringe (Jiangnan, Anhui, China) and 0.45 μm pore-sized polytetrafluoroethylene~~ ~~(PTFE) filter (Jingteng, Tianjin, China) (Chen et al., 2019a). Prior to analysis, filtrates were stored in~~ ~~pre-baked (4 hours at 450 °C) glass vials in a freezer. For each sample, we used a total organic carbon~~ ~~analyzer (Aurora 1030W, OI Analytical, TX, USA) to measure the concentration of WSOC;~~ ~~measurement detection limits and relative standard deviations were 2 μg L−1 and 1 %, respectively.~~ ~~Blank-corrected concentrations are provided in Table S1.~~ All collected snow samples were stored in a freezer at −20 °C until ~~analyzed~~analysis in the laboratory. In the lab process, the samples were firstl~~y~~ melted at room temperature (25 °C). Then 30 mL meltwater was taken for each sample with the clean disposable syringe (Jiangnan, Anhui, China) and injected into the pre-baked (4h, 450 °C) glass bottle passing through 0.45 μm pore-sized polytetrafluoroethylene filters (Jinteng, Tianjin, China). Finally, the concentration of WSOC was measured by the total organic carbon analyzer (Aurora 1030W, OI Analytical, TX, USA), and measurement detection limits and relative standard deviations were 2 μg L^{-1} and 1 %, respectively. ~~Additionally, a blank sample prepared with ultrapure water was measured for~~ ~~blank correction before the sample measurement. The blank concentration of WSOC is 0.35 mg L^{-1} and~~ ~~the blank-corrected WSOC concentrations are provided in Table S1.~~The concentration of WSOC for ultrapure water blank is 0.35 mg L^{-1}, and the value of each sample after blank subtraction is presented in Table S1.

We used 0.4 μm pore-sized polycarbonate filter membranes (Whatman, USA) to isolate BC and other insoluble particles, following the protocols outlined by ~~Shi et al. (2020) and Wang et al. (2014b)~~Wang et al. (2020), after which we employed a custom-developed two-sphere integrating-sandwich (TSI) filter-

1 based spectrophotometer to measure particle absorption. Coupled with the mass of filtered meltwater,

2 these optical measurements were then converted to snowpack BC concentrations. To make these

3 calculations, we applied a BC mass-absorption coefficient (MAC) and absorption Ångström exponent

4 (AAE) of 6.3 m^2 g^{-1} (550 nm) and 1.1, respectively, after Pu et al. (2017). We note that TSI provides

5 greater accuracy and smaller overall uncertainties in the quantification of seasonal snow BC than do

6 thermo-optical carbon analysis (Wang et al., 2020), and thus it has been applied widely in this type of

7 research (Shi et al., 2020). For more detailed information, ~~we refer the reader to~~see Wang et al. (2013b).

8 **2.3 Fluorescence and absorption measurement**

9 We obtained absorbance and fluorescence EEMs for filtered meltwater samples via synchronous

10 absorption-3D Fluorescence scanning spectrometry (Aqualog, Horiba Scientific) with the following

11 measurement parameters: fluorescence spectra excitation range = 240–800 nm in 3 nm intervals,~~;~~

12 emission range = 152.25–929.92 nm in 5.04 nm (8 pixels) intervals, scanning interval = 0.3 seconds.

13 Prior to sample measurement, we analyzed~~analysed~~ aliquots of filtered ultra-pure water (18.2 M ω cm,

14 Milli-q Purification System, Millipore) as analytical blanks. We normalized the fluorescence intensity to

15 that of the water Raman unit (RU), which exhibits a peak excitation wavelength of 350 nm, and deducted

16 this Raman signal from all subsequent sample tests (Lawaetz and Stedmon, 2009). ~~The inner filtration~~

17 ~~effect and Rayleigh scattering peaks were also dispelled following the methods reported by Kothawala~~

18 ~~et al. (2013) and Bahram et al. (2006), respectively.~~ The inner filtration effect correction is based on the

19 absorbance-based approach (Kothawala et al., 2013), using the measured absorbance at each pair of

20 excitation and emission wavelengths across the EEMs to convert the observed fluorescence intensity into

21 the corrected fluorescence intensity. Rayleigh scattering peaks were processed by interpolation algorithm

22 in the EEMscat MATLAB toolbox (Bahram et al. 2006). As fluorescence spectra with wavelengths

1 greater than 600 nm are primarily noise (Zhou et al., 2019b), they are not considered further in this study.

2 ~~Likewise, any samples with absorption spectra of >600 nm wavelengths were subtracted for the baseline~~

3 ~~correction (Chen et al., 2019b).~~

4 We used version 0.6.3 of the MATLAB drEEM toolbox (http://dreem.openfluor.org/; Murphy et al.,

5 2013) to perform PARAFAC analysis on EEMs. Comprising the consistency index, residuals, and visual

6 inspections, the 3-component model is considered more reliable and representative than ~~are~~ the 2–7-

7 component models (Fig. S1 ~~in the Supplement~~) and passes the S4C6T3 split scheme (Fig. S2; Murphy

8 et al., 2013). The contributions of these three components to the overall fluorescence signal are expressed

9 as relative percentages of F_{max} in RU, and the total fluorescence volume (TFV; RU nm^2) is calculated

10 from the EEMs (Song et al., 2019). Normalized TFV equates to NFV (RU nm^2 $(mg\ L^{-1})^{-1}$), TFV

11 $c(WSOC)^{-1}$), where c(WSOC) is the concentration of WSOC in the snow (mg L^{-1})), and represents a

12 sample's fluorescence ability (Chen et al., 2019a).

13 We calculated three fluorescence-derived indices—the fluorescence index (FI), biological index (BIX),

14 and humification index (HIX)—from the ratio of fluorescence intensity at specific excitation and

15 emission wavelengths. As demonstrated by previous studies (Birdwell and Valsaraj, 2010; Huguet et al.,

16 2009; McKnight et al., 2001), these ratios can help characterize potential sources of WSOC. Specifically,

17 the FI is taken to represent the relative amount of DOM derived from terrestrial and microbial/algae

18 sources (McKnight et al., 2001); high values correspond to terrestrially derived organics, and low values

19 reflect microbial sources. The HIX describes the degree of humification of soluble organic matter

20 (Zsolnay et al., 1999). During humification, the aromaticity of organic matter increases as microbial

21 availability decreases, such that higher HIX values correspond to more strongly humified and/or higher

22 aromaticity organics (principally of terrestrial origin), whereas lower values indicate autochthonous or

1   microbial origins. As a measure of autochthonous productivity (Huguet et al., 2009), elevated BIX values

2   are associated with increased contributions of microbial-derived fluorescent organic matter. The three

3   indices are calculated by the following equations (Feng et al., 2016; Huguet et al., 2009; McKnight et al.,

4   2001; Ohno, 2002~~; Huguet et al., 2009; McKnight et al., 2001; Feng et al., 2016~~):

5    $\text{FI} = \dfrac{I(\text{Ex} = 370,\ \text{Em} = 470)}{I(\text{Ex} = 370,\ \text{Em} = 520)},$ (1)

6    $\text{BIX} = \dfrac{I(\text{Ex} = 310,\ \text{Em} = 380)}{I(\text{Ex} = 310,\ \text{Em} = 430)},$ (2)

7    $\text{HIX} = \dfrac{I(\text{Ex} = 254,\ \text{Em} = 435\text{ - }480)}{I(\text{Ex} = 254,\ \text{Em} = 300\text{ - }345) + I(\text{Ex} = 254,\ \text{Em} = 435\text{ - }480)},$ (3)

8   where I is the fluorescence intensity, and Ex and Em represent the excitation and emission wavelengths,

9   respectively. To ensure a direct comparison with prior results, we recalculated published HIX data using

10   the same calculation methods as in our ~~own~~ analyses. According to a previous study, FI values of ≤1.4

11   correspond to terrestrial sources, and values of ≥1.9 denote a primarily microbial origin. The values of

12   1.4–1.9 suggest a mixed origin (McKnight et al., 2001).

13   The absorption spectra of WSOC were derived from 240 to 800 nm in 3 nm intervals. The baseline shifts

14   and scattering effects of the measurement for the absorption spectra were corrected by subtracting the

15   average absorbance above 600 nm from the whole spectrum (Chen et al., 2019b). We converted sample

16   absorbance to an absorption coefficient using the following equation:

17    $a_{\text{WSOC}}(\lambda)~~(\lambda)~~ = \ln(10) \cdot \text{Abs}(\lambda)~~(\lambda)~~ \cdot L^{-1},$ (4)

18   where Abs is absorbance, λ is the wavelength, L is the path length of the cuvette (0.01 m), and a_{WSOC} is

19   the absorption coefficient (m^{-1}).

20   Owing to the absorption characteristics of WSOC, we selected the absorption coefficient at 280 nm

21   ($~~a_{\text{WSOC}}~~a_{\text{WSOC}}(280)$) to characterize the absorption intensity of WSOC for comparison with other studies

22   (Zhang et al., 2010).

To investigate the wavelength dependence of WSOC absorption, we obtained the Absorption Ångström

exponent (AAE) via the following equation (Doherty et al., 2010; Niu et al., 2018; Wang et al., 2013b;

Yan et al., 2016):

$$a_{WSOC}(\lambda)(\lambda) = K \cdot \lambda^{--AAE}, \tag{5}$$

where K is a constant related to WSOC concentration.

We calculated the mass absorption coefficient (MAC$_\lambda$, m^2 g^{-1}) of our samples using the equation (Chen

et al., 2019b; Yan et al., 2016):

$$MAC_\lambda = a_{WSOC}(\lambda)(\lambda) / c(WSOC), \tag{6}$$

where a_{WSOC} is the absorption coefficient derived from Equation (4) and c(WSOC) (mg L^{-1}) is the

concentration of WSOC.

2.4 Snow albedo modeling and radiative forcing calculations

To revealestablish the radiative effect impact of snowpack WSOC in northeastern China, we used

SAMDS to simulate spectral snow albedo. This model is based on the asymptotic radiative transfer theory,

which has been verified by in previous studies (Li et al., 2021b; Wang et al., 2017). and described in

detail by Wang et al. (2017), and it involves parameters including solar zenith angle, impurity

concentrations, snow radius, and equivalent particle size. As described in detail by Wang et al. (2017),

the model involves parameters including solar zenith angle, impurity concentrations, the mass absorption

coefficient of impurities, and snow grain radius. Measured values include the concentration of BC and

the absorption coefficients of WSOC. To quantify the influence of pollutants on snow albedo, we

assumed a semi-infinite snow layer and a uniform snow grain radiusradii of 100 μm for fresh snow and

1000 μm for old snow, consistent with previous studies (Pu et al., 2021). With the solar zenith angle

fixed at 60°, consistent within line with our sampling dates and locations, we calculated the reduction

1   ($\Delta\alpha_i$, i represents BC only, WSOC only, and BC + WSOC, similarly hereinafter.) in spectral snow albedo

2   derived from different types of impurities for the UV–vis (280–400 nm) and ultraviolet–near infrared

3   (UV–NIR; 280–1500 nm) bands. ~~Radiative forcing was then derived by multiplying the albedo reduction~~

4   ~~value by the incident solar radiation (Painter et al., 2013), permitting us to evaluate radiative forcing~~

5   ~~under four scenarios: pure snow, WSOC only, BC only, and WSOC + BC.~~ The radiative forcing resulting

6   from either BC or WSOC in snow (RF_i) was then derived by multiplying the albedo reduction value by

7   the incident solar radiation (Painter et al., 2013):

8     $$RF_i = E \cdot (\alpha_{pure} - \alpha_i) = E \cdot \Delta\alpha_i, \tag{7}$$

9   where α_{pure} is snow albedo for pure snow and E is the average daily downward shortwave solar radiation

10   flux acquired from NASA's Clouds and the Earth's Radiant Energy System (CERES) product "CERES

11   SYN1deg" (https://ceres.larc.nasa.gov/products. php?product=SYN1deg).

12   **3   Results and discussion**

13   **3.1   Characteristics of chemical species**

14   Figure 2a shows the spatial distribution of measured WSOC in seasonal snow across northeastern China.

15   ~~Averaged across our entire study area, t~~The ~~averaged~~mean WSOC concentration (arithmetic mean ±

16   standard deviation) is ~~3.59~~3.6 ± ~~3.19~~3.2 μg g^{-1}, with a maximum of ~~17.99~~18.0 μg g^{-1} and a minimum of

17   ~~0.29~~0.3 μg g^{-1}. Among all~~the~~ five regions, WSOC concentrations are highest in SNC (average 5.7 ±

18   ~~3.68~~3.7 μg g^{-1}), likely reflecting the greater degree of agricultural and industrial activity there compared

19   with other regions (Lu et al., 2011; Wang et al., 2013b). We highlight that both agricultural and industrial

20   sources are considered anthropogenic. In contrast, the ~~our~~ second highest measured concentrations

21   (~~3.35~~3.4 ± ~~1.49~~1.5 μg g^{-1}) are from SEIM, where desertification occurs (Fang et al., 2007) and is

1   therefore considered a natural source of WSOC. For most sites, the underlying surface is desert (Fig. 1a)

2   that was incompletely covered by seasonal snow during the sampling period (Fig. 1d). Consequently, the

3   exposure of natural sandy soils is a potentially significant contributor ~~of~~ to WSOC through aeolian

4   erosion and dry deposition. In NNC, where both the population density and agricultural intensity are

5   lower than in SNC (Choi et al., 2020), the contribution of anthropogenic pollution to the snowpack is

6   correspondingly lower, resulting in a moderate WSOC concentration of 2.7 ± 0.8 µg g^{-1}. Meanwhile, far

7   from intensive human activity, both CBM and NEIM (Fig. 1a, b, and g) returned low WSOC

8   concentrations (CBM: 2.0 ± 1.3 µg g^{-1}; NEIM: 0.5 ± 0.2 µg g^{-1}). Nonetheless, the higher value for CBM

9   betrays the influence of far-traveled anthropogenic pollutants (Wang et al., 2015; Wu et al., 2020; Zhang

10   et al., 2013).

11   ~~In NNC, where both the population density and agricultural intensity are lower than in SNC (Choi et al.,~~

12   ~~2020), the contribution of anthropogenic pollution to snowpack is correspondingly lower, resulting in a~~

13   ~~moderate WSOC concentration of 2.7 ± 0.8 µg g⁻¹. Meanwhile, far from intensive human activity, both~~

14   ~~CBM and NEIM (Fig. 1a, b, and g) returned low WSOC concentrations (CBM: 2.0 ± 1.3 µg g⁻¹; NEIM:~~

15   ~~0.5 ± 0.2 µg g⁻¹). Nonetheless, the higher value for CBM betrays the influence of far-traveled~~

16   ~~anthropogenic pollutants (Wang et al., 2015; Wu et al., 2020; Zhang et al., 2013).~~

Figure 2: Spatial distributions of concentrations of (a) WSOC and (b) BC in snow samples. Sampling sites are divided into the five groups defined in Figure 1. Bubble sizes are proportional to concentrations of WSOC and BC.

In comparison with previous studies, we observed that, with the exception of NEIM, our measured WSOC concentrations are significantly higher than those reported for snow/ice from the Tibetan Plateau (TGL; ~0.71–1.02 µg g^{-1}; Feng et al., 2016), the Alps (~0.14–0.78 µg g^{-1}; Vione et al., 2021), North America (~0.1–0.3 µg g^{-1}; Fellman et al., 2015), and polar regions (~0.12–0.27 µg g^{-1}; Antony et al., 2014), but comparable to values in Laohugou glacier ice from the Tibetan Plateau (~1.02–2.6 µg g^{-1}; Feng et al., 2016~~2018~~, 2018 ~~2016~~) and seasonal snowpack in northwestern China (0.48–7.07 µg g^{-1}; Zhou et al., 2021). This finding implies that snowpack WSOC in northeastern China is significantly contributing ~~significantly~~ to regional and global climate change (Domine, 2002).

A similar spatial pattern is exhibited by snowpack BC (Fig. 2b). For example, of all five regions, the regional mean BC concentration is highest for SNC (~~mean:~~ 922.99923 ± 512.10512 ng g^{-1}), followed by SEIM (659.17659 ± 581.85582 ng g^{-1}), NNC (494.13494 ± 223.81224 ng g^{-1}), and the CBM (391.38391 ± 312.49312 ng g^{-1}). BC concentrations are lowest in NEIM (59.7960 ± 18.6819 ng g^{-1}), in agreement with the values in remote areas reported by Doherty et al. (2010).

3.2 Fluorescence characteristics of WSOC

Three fluorescent components (C1–C3) were captured by resolving the EEM~~s spectra~~s; all fluorescence information is summarized in Table S2. C1 exhibits a primary peak at Ex = 240 nm, Em = 448 nm, indicating the~~a~~ high-oxygenated ~~humic-like substance (~~HULIS~~)~~ found primarily in aromatic conjugated macromolecules (Chen et al., 2016). The weaker secondary peak occurs at longer excitation wavelengths (Ex / Em = 308 / 448 nm), implying a higher aromatic content and greater molecular weight (Coble et al., 1998). Wen et al. (2021) concluded that C1 is probably derived from natural terrestrial sources, such

1 as dust and soil, as proposed originally by Stedmon et al. (2003) and Osburn et al. (2016). Accordingly,

2 we classified C1 as a terrestrial, humic-like substance, hereafter referred to as HULIS-1.

3 C2 exhibits a primary (secondary) peak at Ex = 240 (293) nm, Em = 398 nm, suggestive of the

4 lower-oxygenated HULIS (Chen et al., 2016). Observed in a variety of sources, Stedmon et al. (2003)

5 reported this component in terrestrial end-member samples. Whereas, both Murphy et al. (2011) and

6 Osburn et al. (2016) have since linked C2 to anthropogenic sources, such as urban runoff and sewage.

7 Microbial activity and the degradation of phytoplankton in natural aquatic systems are also thought to

8 contribute to this component (Yamashita et al., 2008; Zhang et al., 2009). Accordingly, we classified C2

9 as humic-like substances with complex origins in terrestrial, anthropogenic, and/or microbial sources,

10 hereafter termed HULIS-2. Unlike HULIS-1 and HULIS-2, C3 is recognizable as a UVB-like protein or

11 tyrosine-like fluorescence (hereafter PRLIS) with a primary (secondary) peak at Ex = 240 (293) nm, Em

12 = 398 nm (Osburn et al., 2016; Stedmon and Markager, 2005). PRLIS reflects autochthonously labile

13 DOM produced by biological processes (Stedmon et al., 2003) and has been reported in previous studies

14 of seasonal snow (Zhou et al., 2019b).

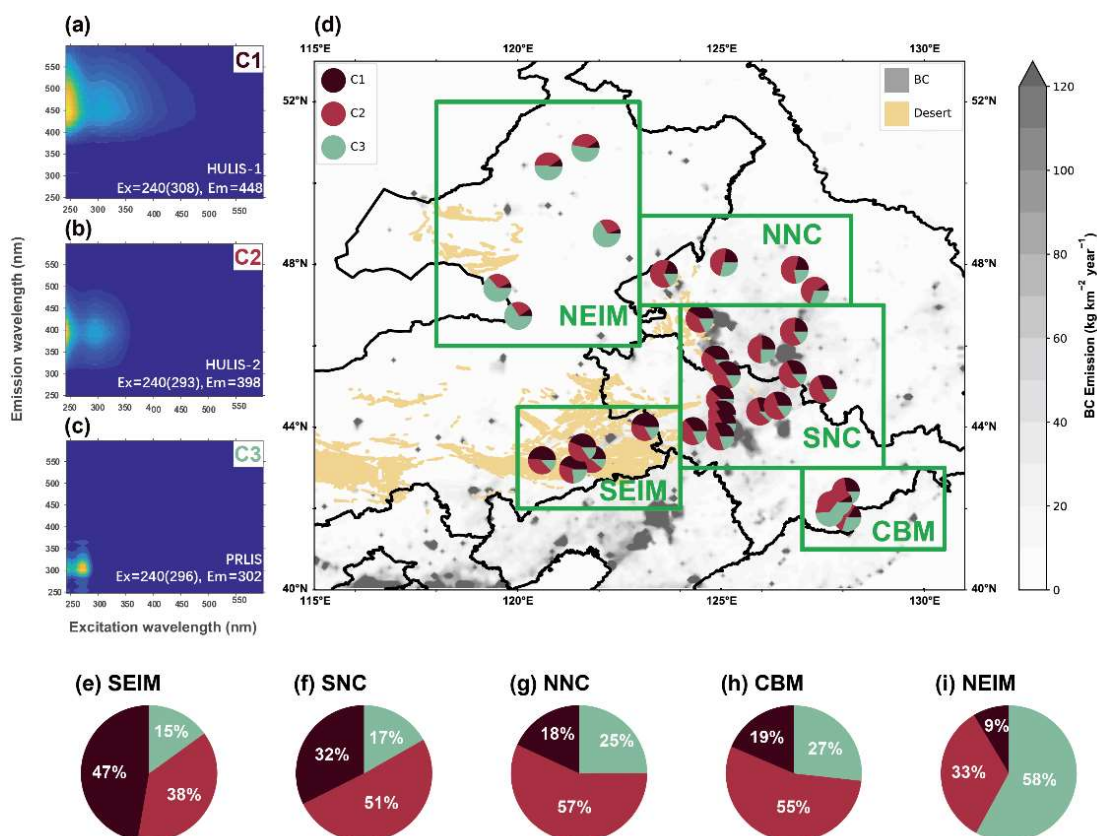



Figure 3: (a–c) Three fluorescent components identified by PARAFAC analysis. (d) Relative contributions of the three components to total fluorescence at each site. HULIS-1, HULIS-2, and PRLIS are represented by the specific colors shown in the legend (top left corner). The distributions of BC emissions and desert areas in our study area are indicated by gray and light yellow, respectively, with darker gray colors indicating higher black carbon concentrations. (e–i) Average contributions of the three components in different groups of samples. BC emission data are derived from the research group at Peking University (http://inventory.pku.edu.cn/ home.html, Wang et al., 2014a); the Chinese desert (sand) distribution dataset is provided by the National Tibetan Plateau Data Center (http://poles.tpdc.ac.cn/zh-hans/data/122c9ac2-53ee-4b9a-ae87-1a980b131c9b/; Wang et al., 2013a).

Figure 3d depicts the spatial distribution of the relative contribution of three components to fluorescence, with the regional averages given in Figure 3e–g. In SEIM, the greatest contribution is that of HULIS-1 (47 %), followed by HULIS-2 (38 %) and PRLIS (15 %), indicating that the signal is dominated by local soil/dust sources, consistent with the local environment (Figs. 2 and 3d). HULIS-2 plays a greater role in SNC, where it accounts for half of the total fluorescence signals,~~; of the remaining half,~~with HULIS-1 is ~~being next~~ most important. This difference in key components between SEIM and SNC illustrates the change in the primary source of fluorescence intensity. ~~Indeed, with the most intensive human activity~~

(e.g., agriculture, industrial emissions) being located in SNC (Figs. 1a and 3d; Guo and Hu, 2022), HULIS-2 might be derived from any combination of terrestrial, anthropogenic, and microbial sources. Nonetheless, in agreement with previous studies (Zhou et al., 2019b), our combined analysis suggests that anthropogenic activity is the main contributor to seasonal snow in northeastern China. Indeed, although HULIS-2 might be derived from any combination of terrestrial, anthropogenic, and microbial sources (Osburn et al., 2016; Stedmon et al., 2003; Yamashita et al., 2008; Zhang et al., 2009), human activity (e.g., agriculture, industrial emissions) is most intensive in SNC. Therefore, our combined analysis suggests that anthropogenic source is the main contributor to seasonal snow in SNC (Figs. 1a and 3d; Guo and Hu, 2022). The conclusion is also in good agreement with the previous study (Zhou et al., 2019b). As in SNC, HULIS-2 also represents approximately half of the fluorescence signal in both NNC and the CBM. The background environment of NNC is similar to that of SNC, with dense urban cities and populations. In the latter CBM, which is although heavily forested (Fig. 1a; Guo and Hu, 2022), the long-range transport of anthropogenic pollutants is responsible for the dominance of HULIS-2, as discussed in Sect. 3.1. HULIS-1 accounts for less than PRLIS in both NNC and the CBM, which we posit reflects the concealment of bare soil surfaces by deep snow and the importance of biological processes due to the heavy vegetation cover. PRLIS accounts for >50 % of the total fluorescence signal in NEIM, followed by HULIS-2; HULIS-1 contributes relatively little in this region. We attribute this pattern to both the extensive grassland and forest cover, which obscures bare soil surfaces, and the distance from anthropogenic pollution. These together serve to amplify the importance of biological processes (Zhou et al., 2019a). Taken as a whole, the respective contributions of HULIS-1, HULIS-2, and PRLIS to the fluorescence signals in our study area are ~30 %, ~50 %, and ~20 %, respectively. We note that these findings correspond well with the background environmental conditions.

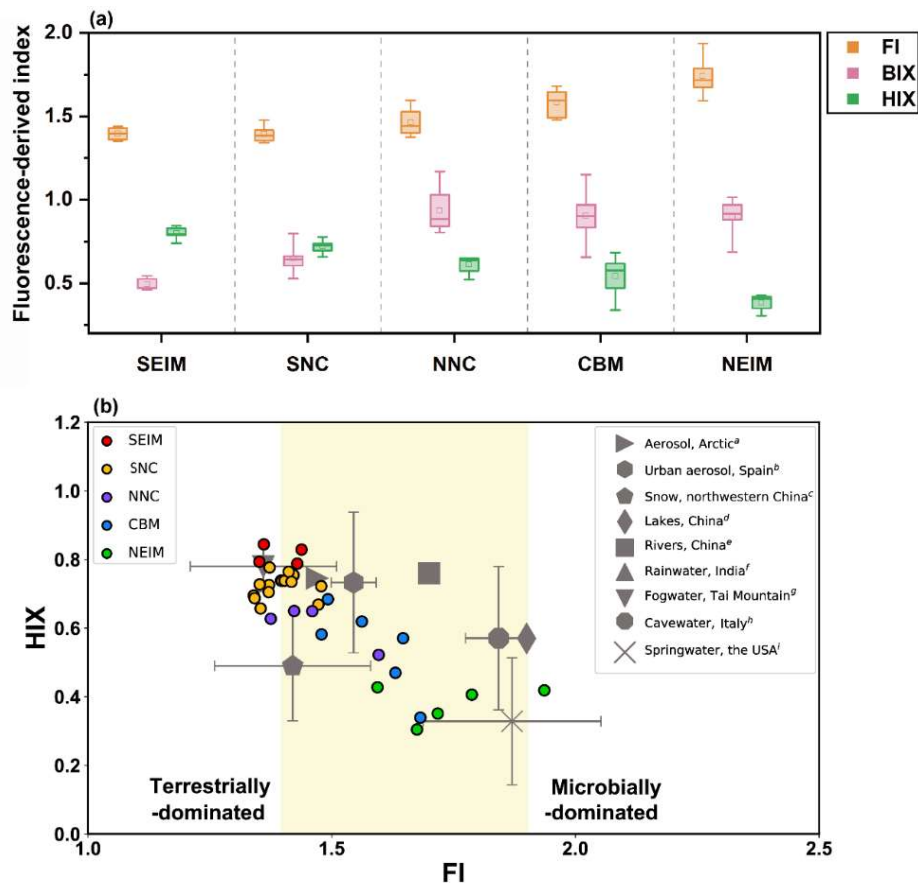



Figure 4: (a) Variations in fluorescence-derived indices among the five groups. Boxes denote the 25th and 75th quantiles, and horizontal lines represent median values. Averages are shown as small boxes, the whiskers denoting maximum and minimum data. (b) Comparison plots of HIX versus FI for the seasonal snow surface samples (colored dots) from northeastern China, together with the average and standard deviation of different types of WSOC (grey markers) adapted from: Arctic aerosols ([a] Fu et al., 2015), Spanish urban aerosols ([b] Mladenov et al., 2011), seasonal snowpack in northwestern China ([c] Zhou et al., 2019b), Chinese lakes and rivers ([d, e] Zhou et al., 2017), rainwater from Rameswaram, India ([f] Salve et al., 2012), fog water from Tai Mountain, China ([g] Birdwell and Valsaraj, 2010), cave water from Frasassi Caves, Italy ([h] Birdwell and Engel, 2010), and spring water in the USA ([i] Birdwell and Engel, 2010). Shaded areas represent mixed WSOC signatures.

The FI, BIX, and HIX indices reveal spatial variability in fluorescence characteristics and thus permit the tracing of potential sources. Regionally averaged FI, BIX, and HIX values are depicted in Figure 4a.

Our results show that, ~~in general,~~ the FI values are~~varies in the range of~~ 1.34–1.94 (mean = 1.49), BIX ~~between~~ are 0.46 and 1.17 (mean = 0.74), and HIX ~~between~~ are 0.30 and 0.84 (mean = 0.64). By comparison, reported mean FI, BIX, and HIX values for seasonal snow in Xinjiang (northwestern China) are 1.42, 0.76, and 0.55, respectively (Zhou et al., 2019b), suggesting that the impacts of humification

1   and WSOC aromaticity are slightly ~~higher~~ greater in ~~our~~ the study area than in Xinjiang. This outcome

2   implies a relatively strong terrigenous signal and correspondingly weaker biogenic signal in the seasonal

3   snowpack of northeastern China. Regionally, SEIM exhibits the lowest FI (mean = 1.40) and BIX (mean

4   = 0.49) values but the largest HIX value (mean = 0.80), further confirming the strong influence of highly

5   aromatic, terrestrially derived WSOC in this region relative to the others. In contrast, NEIM returns the

6   highest FI (mean = 1.74) and BIX (mean = 0.89) values, but the lowest HIX value (mean = 0.38),

7   indicating the dominance of low-aromatic WSOC of microbial origin. Intriguingly, our results reveal that

8   FI and BIX rise ~~monotonously~~generally with decreasing (increasing) fractional contributions of HULIS-

9   1 (PRLIS), whereas HIX exhibits a similar but contrasting pattern. Together, the comprehensive dataset

10   described above verifies the regional variability in the terrestrial contributions to WSOC, with~~in which~~

11   SEIM > SNC > NNC > CBM > NEIM; this pattern is reversed for microbially sourced WSOC.

12   Figure 4b illustrates HIX versus FI as a scatterplot, compared with published data for different sample

13   types; the shaded area depicts the region in which the FI value is >1.4 but <1.9. ~~As FI values of ≤1.4~~

14   ~~correspond to terrestrial sources and values of ≥1.9 denote a primarily microbial origin, values of 1.4–~~

15   ~~1.9 suggest a mixed origin.~~ In general, FI exhibits a rising trend with declining HIX across northeastern

16   China. For both SEIM and SNC, FI occupies a narrow range centered on 1.4, indicating either a

17   predominantly terrestrial or mixed origin. We note that these results are comparable to those of fog water

18   from the Tai Mountain, Arctic atmospheric aerosols, and seasonal snow in northwestern China (Birdwell

19   and Valsaraj, 2010; Fu et al., 2015; Zhou et al., 2019b). Further, we highlight that HIX values are

20   marginally higher in SEIM than elsewhere, suggesting a stronger influence from highly humified WSOC

21   that probably reflects the extensive deserts and exposed earth in this region. FI values for NNC and the

22   CBM fall within the range of 1.4–1.7 and thus reflect a mixed origin, in line with previous data from

urban aerosols in Spain and Chinese river water samples (Mladenov et al., 2011, Zhou et al., 2017).

When combined, the FI and HIX values for NNC and CBM snowpack indicate that WSOC in these

regions bears a stronger terrestrial signature than do water samples from Chinese lakes and Italian caves

(Birdwell and Engel, 2010; Zhou et al., 2017). Finally, FI values for NEIM fall within a range of 1.6–

2.0, comparable to values ~~from~~ for spring water in the USA (Birdwell and Engel, 2010), thus implying a

predominantly microbial or mixed origin.

3.3   Comparisons of fluorescence and absorption characteristics

~~Figure 5a depicts TFV as a measure of the spatial distribution of absolute WSOC fluorescence intensity in the snowpack of northeastern China; aWSOC(280) is shown in Figure 5b for comparison.~~ Figure 5a describes the spatial distribution of $a_{WSOC}(280)$ as WSOC absorption in the snowpack of northeastern China; Figure 5b depicts TFV as a measure of the spatial distribution of absolute WSOC fluorescence intensity for comparison. In general, TFV and ~~awsoc~~$a_{WSOC}(280)$ both exhibit large spatial variability in

the range of 690–18600 RU nm^2 and 0.42–16.98 m^{-1}, respectively. Regional mean values are 7700 ±

2800 RU·nm^2 (TFV) and 6.90 ± 2.39 m^{-1} (~~awsoc~~$a_{WSOC}(280)$) for SEIM, 12400 ± 4300 RU·nm^2 (TFV)

and 11.48 ± 3.96 m^{-1} (~~awsoc~~$a_{WSOC}(280)$) for SNC, 7800 ± 500 RU·nm^2 (TFV) and 7.02 ± 0.88 m^{-1}

(~~awsoc~~$a_{WSOC}(280)$) for NNC, 3900 ± 2500 RU·nm^2 (TFV) and 3.97 ± 2.46 m^{-1} (~~awsoc~~$a_{WSOC}(280)$) for

the CBM, and 1000 ± 300 RU·nm^2 (TFV) and 0.83 ± 0.23 m^{-1} (~~awsoc~~$a_{WSOC}(280)$) for NEIM. We note

that both distributions are consistent in space (Fig. 5e), with the highest concentrations in SNC and the

lowest in NEIM. Moreover, the ~~awsoc~~$a_{WSOC}(280)$ value for SNC is an order of magnitude larger than

that for NEIM, implying that the impact of WSOC on snow albedo at UV wavelengths is significant in

SNC but less notable in NEIM in general (see Sect. 3.5). Previous work has reported a similarly broad

range of snowpack ~~awsoc~~$a_{WSOC}(280)$ (0.15–10.57 m^{-1}) in northwestern China (Zhou et al., 2019b).

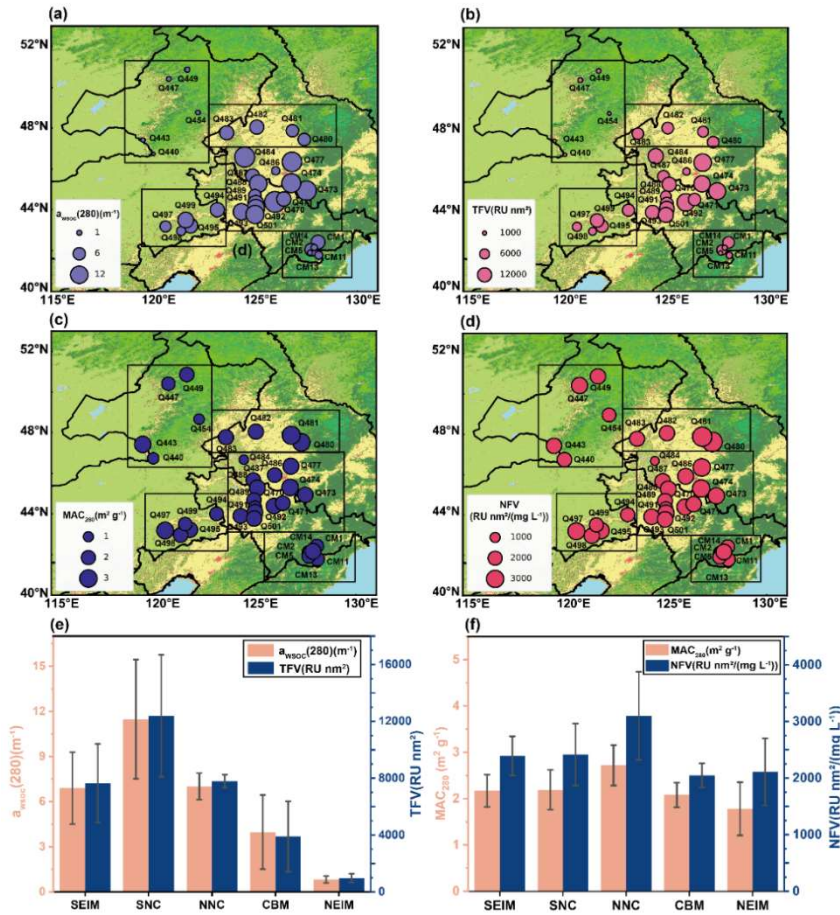



Figure 5: Spatial distribution of (a) ~~awsoc~~a_{WSOC}**(280) (m⁻¹), (b) TFV (RU nm²)~~,~~ (c) MAC$_{280}$ (m² g⁻¹) and (d) NFV (RU nm² (mg L⁻¹)⁻¹). Regional averages for (e)** ~~awsoc~~a_{WSOC}**(280), TFV, and (f) MAC$_{280}$~~, and~~ NFV for the five groups. Error bars in (e) and (f) represent the standard deviations of** ~~awsoc~~a_{WSOC}**(280), MAC$_{280}$, TFV, and NFV for the five groups, respectively.**

Two additional fluorescence and absorption capacity indices, identified as NFV and MAC$_{280}$, are proven tools for revealing WSOC's fluorescence and absorption characteristics and they are related to chemical composition, structure, and source (Chen et al., 2019a). For the present ~~our~~ study area as a whole, mean NFV and MAC$_{280}$ values are ~~2411.57~~2412 ± ~~373.63~~ 374 RU nm² (mg L⁻¹)⁻¹ and ~~2.17~~2.2 ± ~~0.49~~0.5 m² g⁻¹, respectively. Both indices exhibit a narrow range, with regional means ranging from 2100 ± 600 to 3100 ± 800 RU nm² (mg L⁻¹)⁻¹ and from ~~1.78~~1.8 ± ~~0.57~~0.6 to ~~2.72~~2.7 ± ~~0.44~~0.4 m² g⁻¹, respectively, in contrast to the broad inter-regional disparities in TFV and ~~awsoc~~ a_{WSOC}(280). Moreover, the spatial patterns of NFV and MAC$_{280}$ are similar, with the highest values in NNC. We speculate that this result

reflects the comparatively high low-oxygenated HULIS-2 fraction measured in the NNC samples (Fig.

3g), as the lower-oxygenated BrC (e.g., HULIS-2) has a higher absorption capacity (Browne et al., 2019).

Scatterplots ~~for~~ of a_{WSOC}(280) versus~~with~~ TFV, F_{max}(HULIS-1), F_{max}(HULIS-2), and F_{max}(PRLIS)

are provided in Figure S4 to further demonstrate the close relationship between fluorescence and the

absorption characteristics of WSOC in our snow samples. As samples Q480, Q484, and Q477 deviate

considerably from the respective confidence intervals, we did not include them in our analyses.

Surprisingly, we found that TFV is closely correlated to a_{WSOC}(280), with P < 0.001 and all

datapoints located close to the line of best fit, indicating that the three components (HULIS-1, HULIS-

2, PRLIS) contributing to the total fluorescence are also responsible for the majority of absorption. For

each component, our data show that F_{max}(HULIS-1) is most closely correlated with a_{WSOC}(280),

followed by F_{max}(HULIS-2). The correlation between F_{max}(PRLIS) and a_{WSOC} (280) is the

~~poorest~~weakest, yet it is still significant (P < 0.001). Together, our results imply that HULIS-1 is

probably the greatest contributor to light absorption, with PRLIS being the least important.

3.4   Fractional contributions of different WSOC components to light absorption

~~Previous studies of atmospheric aerosols, water, and snow/glacier ice have typically regarded WSOC as~~

~~a whole when discussing its impact on light absorption (Barrett and Sheesley, 2017; D'Sa et al., 2014;~~

~~Niu et al., 2018; Wu et al., 2019). Yet, depending on environmental conditions, the various components~~

~~of WSOC play measurably different roles in light absorption according to their concentrations and optical~~

~~properties (Zhou et al., 2022). Although the qualitative analysis described above have provided plausible~~

~~information about the component-specific influence of WSOC on light absorption, the quantitative~~

~~fractional contributions of specific components to light absorption are still unknown. Recently, Chen et~~

~~al. (2019a) collected atmospheric aerosol samples in PM$_{2.5}$ over Xi'an, China, and successfully attributed~~

1   ~~the dithiothreitol (DTT) activity levels to various BrC components by coupling DDT and BrC datasets.~~

2   ~~A similar attribution method has been applied to various research areas, including climate change,~~

3   ~~extreme weather, and atmospheric environments (Cao et al., 2015; Pokrovsky, 2019; Xin et al., 2016;~~

4   ~~Zhao et al., 2019).~~ In this study, we applied a multiple linear regression method comparable to that of

5   Chen et al. (2019a) to derive the fractional contribution of each WSOC component to light absorption.

6   ~~We note that, despite this method having been used elsewhere (Wu et al., 2022; Wu et al., 2021), it~~

7   ~~remains a highly innovative approach to evaluating the light absorption of snowpack WSOC.~~ Table S3

8   lists the statistical results of the fitted light absorption coefficient, based on the F_{max} data for three

9   fluorescent components of EEM analysis. As the fitted results can explain ~94 %–99 % of the variance

10 in measured light absorption within the 280–400 nm range, we conclude that the fusion of multiple

11 fluorescent components is an effective means of describing most of the spatial features of WSOC light

12 absorption throughout northeastern China. Accordingly, the wavelength-dependent fractional

13 contributions of each component of light absorption in this band (280–400 nm) are reported in Figure 6.

14 For the~~our~~ entire study area, light absorption is dominated by high-oxygenated HULIS-1, which accounts

15 for ~56 %–65 % of the contribution across UV wavelengths. Further more, with increasing wavelength

16 the contribution of ~~we observed that the~~ HULIS-1 first increases from~~contribution rises slightly from~~

17 280 to ~340 nm, ~~after which there is a~~and then decreas es~~ing trend as wavelength increases~~. In contrast,

18 HULIS-2 exhibits a valley-type pattern in terms of its fractional contribution at wavelengths between

19 280 and 400 nm and is responsible for ~19 %–30 % of all light absorption.

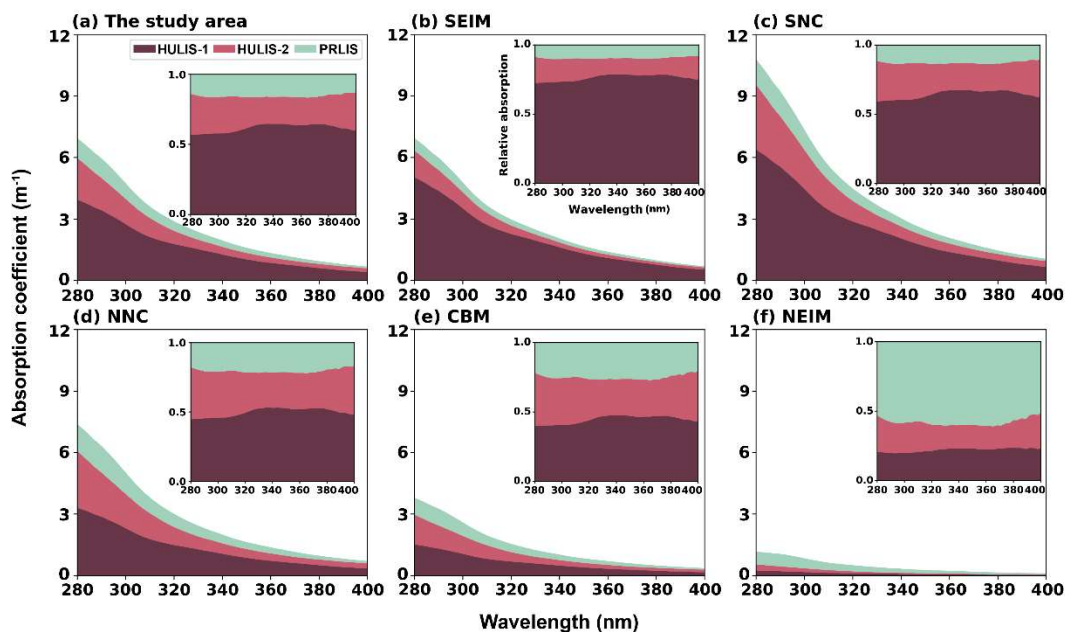



Figure 6: Relative contributions of the three components to the total absorption of samples in (a) the whole study area and (b–f) each of the five groups.

PRLIS contributes the least (~12 %–17 %) to light absorption and exhibits a similar wavelength-dependent pattern to HULIS-1. These results are consistent with the qualitatively comparative analysis described in Sect. 3.3. Previous studies have also highlighted this dominance of high-oxygenated compounds in WSOC light absorption, based on samples impacted by naturally and anthropogenically derived soils (Zhou et al., 2022). Conversely, the total absorption coefficient of WSOC decreases with the increasing wavelength between 280 and 400 nm, in accord with previous studies (Andreae and Gelencser, 2006; Chakrabarty et al., 2010; Gustafsson et al., 2009; Wu et al., 2019). The AAE lies primarily between 5.0 and 8.0 (mean = 6.6) in the range of 280–400 nm, which is in agreement with results from snow collected from the Arctic, the northern Tibetan Plateau, and northwestern China (Voisin et al., 2012; Yan et al., 2016; Zhou et al., 2021).

For each component, the wavelength-dependent variability in light absorption is similar among all five regions, although the magnitude of each contribution varies from region to region. Moreover, compared with the spectral results, we found that the solar-radiation-weighted broadband light absorption iswas a

1   more meaningful parameter for researchers studying climate change and atmospheric radiation.

2   Therefore, the broadband results in Figure 7a for 280–400 nm absorption contributions—HULIS-1

3   (62 %), HULIS-2 (21 %), and PRLIS (17 %)—are average values for the whole study area. On a regional

4   scale, the HULIS-1 contribution to light absorption (280–400 nm) follows the spatial pattern SEIM >

5   SNC > NNC > CBM > NEIM. We note that HULIS-1 dominates light absorption in SEIM, SNC, and

6   NNC but has a minor impact in NEIM compared with the other two components. In contrast, the impact

7   of HULIS-2 varies only slightly among the five regions, with the greatest contributions in NNC and

8   CMB, and the lowest in NEIM. The contribution of PRLIS is essentially opposite to that of HULIS-1,

9   being dominant in NEIM but of relatively minor importance elsewhere. As shown in Figure 7b, light

10   absorption contributions at 280 nm are consistent with the broadband results (Fig. 7a) in terms of the

11   regional pattern, although specific values differ because of the different wavelength-dependent properties

12   of light absorption for the three WSOC components.

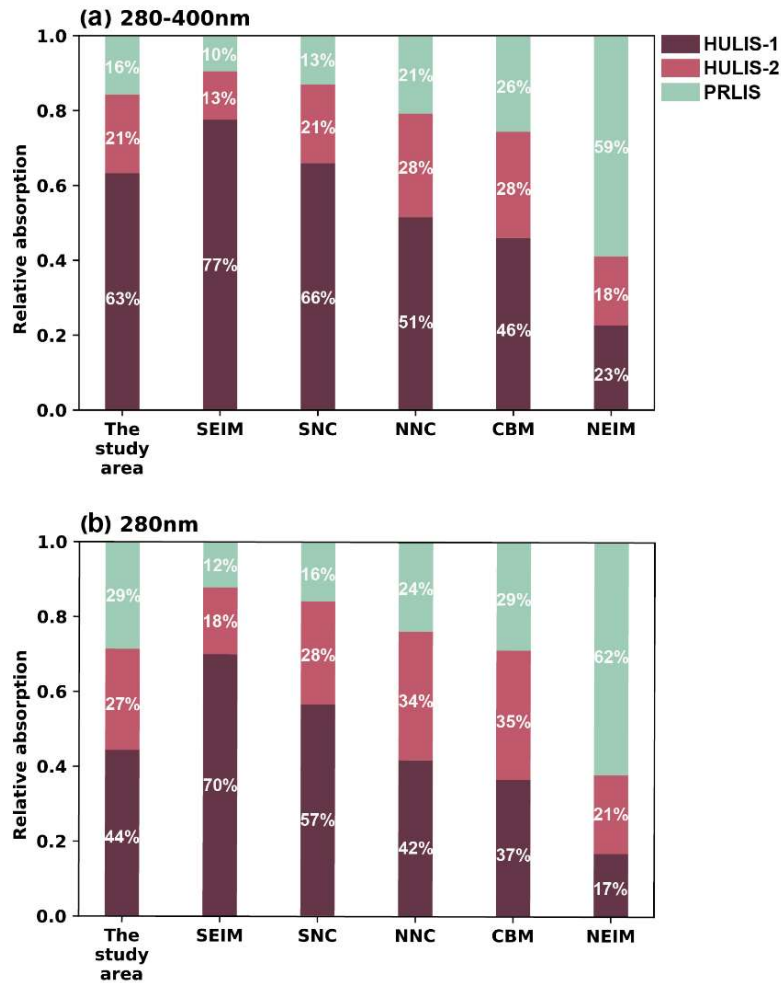



Figure 7: Regional averages for the relative contributions of the three fluorescent components to light absorption at wavelengths of (a) ~~280 nm~~ 280–400 nm **and (b)** ~~280–400 nm~~ 280 nm**.**

We find it noteworthy that, for each component, the overall regional pattern of its contribution to light

absorption aligns with its impact on fluorescence signals, thereby confirming the viability of the

attribution analysis employed in this~~our~~ study. Nonetheless, we observed that the magnitude of each

component's contribution varies relative to its respective fluorescence signal. For instance, HULIS-1

returns a greater contribution to light absorption than its fluorescence signal, in contrast to HULIS-2.

One plausible explanation for this discrepancy is that the apparent fluorescence quantum yields (AQYs~~s~~),

which are essentially the ratio of fluorescence intensity versus absorption intensity, are different for each

component. Indeed, in their comprehensive field-based study of BrC fluorescence and absorption

properties in northern China, Wen et al. (2021) reported that the AQYs of WSOC decreases with

increasing HIX, meaning that components with higher HIX values. Such as HULIS-1, have lower AQYs

than doseHULIS-2. Thus, the contribution of HULIS-1 to the fluorescence signals will be smaller than

its contribution to light absorption, and vice versa for HULIS-2.

3.5 Albedo reduction and radiative forcing attributed to snowpack WSOC

The strong light absorption of WSOC in UV bands has important ramifications for snow albedo and

radiative forcing throughout northeastern China. However, owing to the chemical and optical complexity

of WSOC components, quantitative estimates for snowpack light absorption remain poorly understood.

For example, although prior work in northeastern China has focused on BC (Wang et al., 2013b) and

other water-insoluble light-absorbing particles (Wang et al., 2017; Zhao et al., 2014) via field

measurements, model simulations, and satellite remote sensing (Pu et al., 2019), the specific impacts of

WSOC have not been studied. Consequently, ours is the first study to report on the impact of WSOC on

snow albedo and radiative forcing in northeastern China and to compare these data with BC results to

highlight the non-negligible role of WSOC.

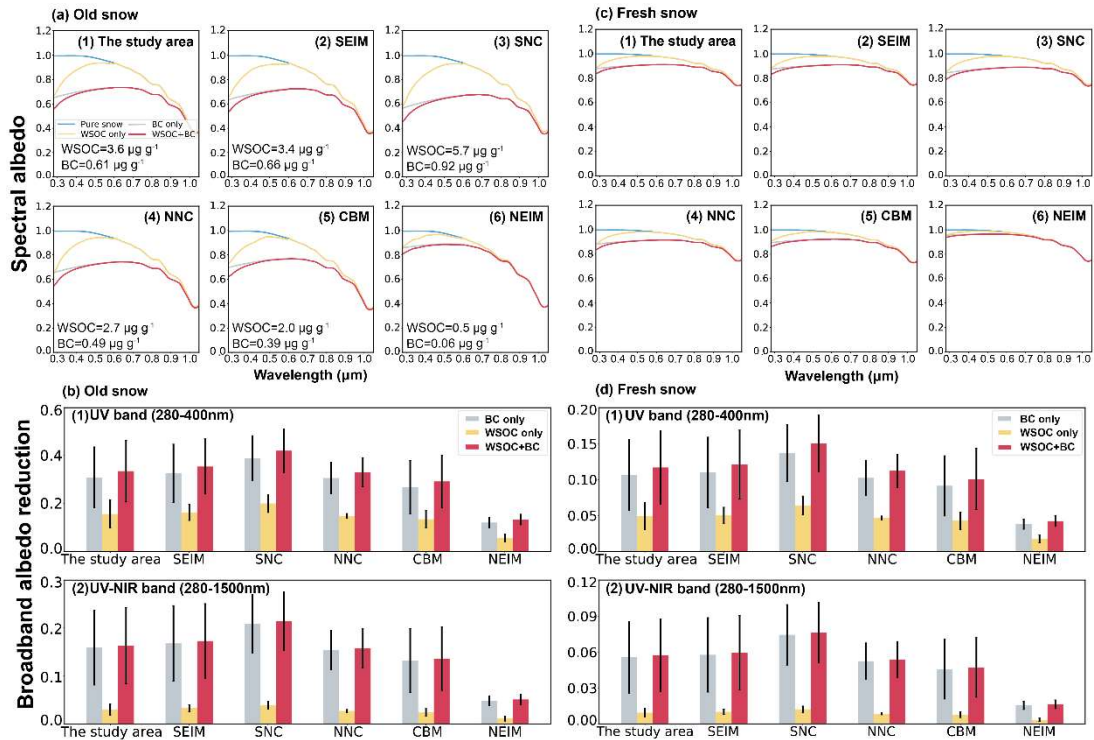



Figure 8: (a) and (b): Simulated snow spectral albedo and broadband albedo reductions—under various contamination scenarios and for different regions—for old snow (radius = 1000 µm). (c), (d) Simulated snow spectral albedo and broadband albedo reductions—under various contamination scenarios and for different regions—for fresh snow (radius = 100 µm). Colors represent the different types of snow (pure snow, BC- or WSOC-contaminated snow, and snow polluted by both WSOC and BC).

Figure 8 shows the regional-mean spectral snow albedo as well as the reduction in albedo due to WSOC, BC, and WSOC + BC. We assume a snow radius of 100 µm for fresh snow and 1000 µm for old snow.

Our findings reveal that WSOC induces a marked decline in albedo within the UV and short-wave VIS bands, with the magnitude of albedo reduction growing rapidly as wavelength shrinks owing to the large AAE value of WSOC. In comparison, BC induces a widespread albedo reduction spanning the UV to NIR bands, and wavelength-dependent variations are significantly smaller than those of WSOC. For VIS and NIR, the reduction in albedo is dominated by BC, whereas the impacts of WSOC and BC are comparable atin UV wavelengths, a pattern that is consistent with the results of studies of atmospheric aerosols (Shamjad et al., 2016). We note that these characteristics persist throughout northeastern China despite regional variability in environmental conditions and snowpack types (old vs. fresh snow).

1 For broadband wavelengths, our results indicate that the WSOC-induced (mean = 3.6 μg g^{-1}) albedo

2 reduction for 280–400 nm wavelengths in old (fresh) snow is 0.16 (0.05) across the whole study area,

3 which corresponds to approximately 5050.3 % (4646.3 %) the impact of BC (mean = 0.6 μg g^{-1}).

4 Regionally, the greatest decline in albedo occurred in SNC, where a mean WSOC of 5.7 μg g^{-1} resulted

5 in a reduction of 0.20 (0.06) in the 280–400 nm range for old (fresh) snow. In contrast, the smallest

6 decline in albedo was observed in NEIM, with reductions of 0.06 (0.02) resulting from an average WSOC

7 concentration of 0.5 μg g^{-1}. Compared with the UV bands, a WSOC-induced albedo reduction of 0.03

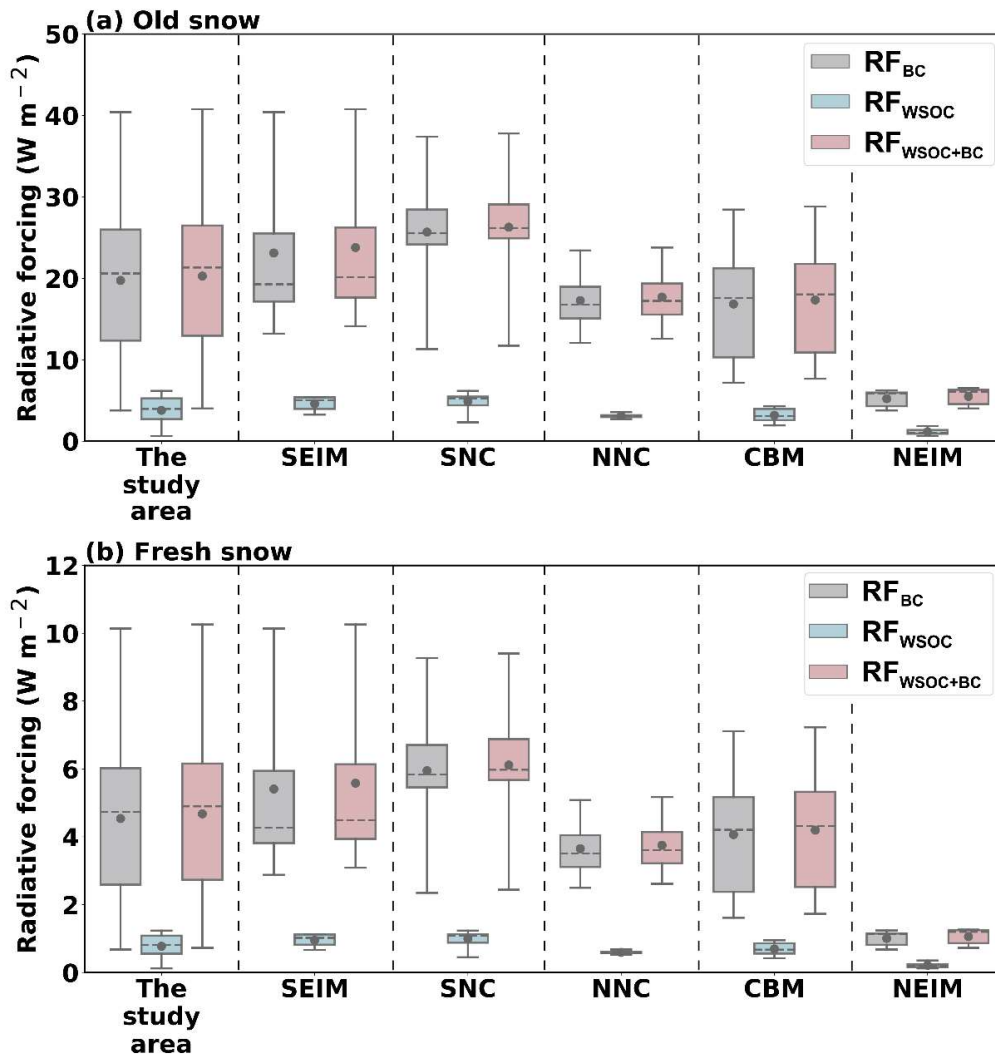
8 (0.009) over the UV–NIR range (280–1500 nm) accounts for only ~1918.8 % (1716.7 %) of that due to

9 BC in theour study area. The regional mean for old (fresh) snow falls in the range of 0.01–0.04 (0.003–

10 0.012), with the highest (lowest) values occurring in SNC (NEIM). However, we observed the highest

11 ratio of WSOC- to BC-induced albedo reduction in NEIM. Together, these results indicate that WSOC

12 plays a potentially important role in altering UV snow albedo in NEIM, despite its relatively low

13 concentrations in the regional snowpack.

2 **Figure 9: Radiative forcing due to different pollutants in (a) old or (b) fresh snow. Gray, blue, and red indicate**

3 **the radiative forcing of BC, WSOC, and BC + WSOC, respectively.**

4 Radiative forcing is an important index that directly reflects the impact of snowpack WSOC on the

5 regional radiation balance and climate (Beres et al., 2020). Previous studies have tended to focus on

6 calculating instantaneous radiative forcing values; however, in reality, time-averaged results are more

7 valuable for climate research. Here, we present data on the daily mean radiative forcing due to WSOC,

8 BC, and WSOC + BC (Fig. 9)~~, calculated using the methodology of Wang et al. (2017)~~. ~~In general, f~~For

9 northeastern China ~~we found~~ the mean radiative forcing of WSOC in old (fresh) snow ~~to be~~is ~~3.78~~3.8

10 (~~0.77~~0.8) W m^{-2}, with regional mean values varying from ~~1.15~~1.2 (~~0.21~~0.2) to ~~4.88~~4.9 (1.0) W m^{-2}. Zhou

et al. (2021) reported daily mean radiative forcing by regional WSOC (0.6–7.1 µg g^{-1}) of between ~0.04

and ~0.6~~0.59~~ W m^{-2} for northwestern China, which is comparable to our values in fresh snow.

Furthermore, the ratio of WSOC-driven to BC-driven radiative forcing varies within the range of

10~~10.3~~ %–32~~.0~~ % (10~~9.8~~ %–31~~30.8~~ %) for old (fresh) snow, which is consistent with the results of our

calculated albedo reductions. These results confirm that the role of WSOC must not be ignored in

discussions about radiative balance in northeastern China. Similarly, the sizeable impact of WSOC on

the absorption of UV radiation has the potential to influence biogeochemistry (Helms et al., 2013; Seekell

et al., 2015)~~, and~~ snow photochemical processes (e.g., photolysis of nitrate (NO$_3^-$) and nitrite (NO$_2^-$) in

snow, in addition to the release of NO$_x$ (NO + NO$_2$ and HONO). Snow photochemistry is beyond the

scope of this study, however, the high concentrations of WSOC and nitrate (not shown) pollution in

northeastern China make this a logical next step for research in this field.

4   Conclusions and atmospheric implications

During 2020 and 2021, we collected 34 surface samples of seasonal snow from sites throughout

northeastern China to investigate the fluorescence characteristics, optical properties, and radiative effects

of snowpack WSOC. With an average concentration of WSOC of ~~3.59~~3.6 ± ~~3.19~~ 3.2 µg g^{-1}, our results

returned regional mean values of ~~3.35~~3.4 ± ~~1.49~~ 1.5 µg g^{-1} (SEIM), ~~5.73~~5.7 ± ~~3.68~~ 3.7 µg g^{-1} (SNC),

~~2.70~~2.7 ± ~~0.75~~0.8 µg g^{-1} (NNC), ~~1.95~~2.0 ± ~~1.28~~1.3 µg g^{-1} (CBM), and ~~0.50~~0.5 ± ~~0.19~~0.2 µg g^{-1} (NEIM),

indicating a considerable degree of regional variability of WSOC mass loadings. Measured values of

WSOC fluorescence intensity (690–18600 RU nm^2) and light absorption (0.4–17~~.0~~ m^{-1}) are also highly

variable.

~~In the first study of its kind~~Moreover, we also used EEMs and PARAFAC to identify three fluorescence

WSOC components prevalent in northeastern China and analyzed their regional differences. ~~Specifically,~~

these include the high-oxygenated HULIS-1, which is a terrigenous, humic-like component, and the low-oxygenated HULIS-2, which is a humic-like component derived from mixed sources, such as anthropogenic activity, microbial processes, and soil. The third component, PRLIS, is a protein-like substance derived from autochthonous biological activity. In SEIM, which is characterized by desert and bare soil surfaces, the signal of high-oxygenated and terrigenous HULIS-1 is dominant (47 %) and HIX values the highest, whereas FI and BIX are the lowest. Together, these findings reveal a The high degree of humification and minimal bioavailability of WSOC, indicating indicate that snowpack WSOC originates primarily from soil sources. In contrast, the autochthonous PRLIS signal (58 %) dominates in remote and clean NEIM, which also exhibits the lowest HIX values and highest FI and BIX values. We propose that the elevated bioavailability of this remote, forested region indicates a predominantly biological origin for NEIM snowpack WSOC. Low-oxygenated and anthropogenic HULIS-2 dominates the densely populated and intensively farmed SNC (51 %) and NNC (57 %) regions, where HIX, FI, and BIX values are moderate, leading us to conclude that the snowpack WSOC is of mixed origin. in SNC and NNC are influenced more by anthropogenic sources. In CBM of the forest environment, the impact of long-distance transport of pollutants is greater than that of the background environment. The above conclusions are also verified by fluorescence-derived indices.

We employed multiple regression analysis to estimate the fractional contributions of different WSOC components to snowpack light absorption. Throughout our study area, HULIS-1 tends to be the greatest contributor (~56 %–65 %) over the 280–400 nm range, followed by HULIS-2 (~19 %–30 %) and PRLIS (~12 %–17 %). On a more regional basis, light absorption remains dominated by HULIS-1 in SEIM, SNC, NNC, and the CBM, whereas PRLIS takes a leading role in NEIM. In contrast to its primary role

in fluorescence, the contribution of HULIS 2 to light absorption is relatively low across all regions, potentially reflecting the variable fluorescence quantum yields (AQYs) of the different components. Finally, we compared the impact on snow albedo and radiative forcing of WSOC relative to BC. With an average concentration of 3.6 μg g^{-1}, our results indicate that WSOC induces an albedo reduction across northeastern China of 0.16 (0.05) in old (fresh) snow over the 280 400 nm range, and thus represents approximately 50 % (46 %) of the albedo reduction due to BC (average concentration = 0.6 μg g^{-1}). We note, however, that the WSOC driven reduction in the UV NIR spectrum (280 1500 nm) is only 0.03 (0.009), corresponding to 19 % (17 %) that of BC. The average radiative forcing of WSOC in old (fresh) snow in northeastern China is 3.8 (0.8) W m^{-2}, equating to 19 % (17 %) of the BC-induced radiative forcing. Ultimately, our findings indicate that WSOC (1) plays an important role in the regional radiation balance and (2) might significantly affect biogeochemical and photochemical processes in snow. We employed multiple regression analysis to estimate the fractional contributions of different WSOC components to snowpack light absorption. Throughout our study area, HULIS-1 tends to be the greatest contributor (~56 %–65 %) over the 280–400 nm range, followed by HULIS-2 (~19 %–30 %) and PRLIS (~12 %–17 %). In contrast to its primary role in fluorescence, the contribution of HULIS-2 to light absorption is relatively low across all regions, potentially reflecting the variable molecular structure of different components. Finally, we highlighted that the average RF due to WSOC in old (fresh) snow in northeastern China is 3.8 (0.8) W m^{-2}, which is equal to 19 % (17 %) of the BC-induced radiative forcing. Therefore, we demonstrated the important impacts of WSOC on the snow energy budget and potentially on triggering snow photochemistry.

We indicate that our study could contribute to the understanding of carbon cycling processes, regional air quality, hydrological processes, and climate change in the earth systems. For example, the abundant

1 ~~WSCO~~ WSOC concentrations measured in this study implied the significant carbon input from the

2 atmosphere to the snowpack through wet or dry depositions in northeastern China. While the complex

3 chemical compositions of snowpack WSOC could further influence the carbon balance of the snow

4 environment by affecting microbial activities (Stedmon et al., 2007). The strong absorption properties of

5 WSOC in the UV–Vis band also implied its important role in initiating snow photochemistry (McNeill

6 et al., 2012). It ~~which~~ will change the composition of organic compounds in the snow in turn (Grannas

7 et al., 2007), and affect the surrounding air quality by releasing oxidizing gas like NO_x into the

8 atmosphere (Zatko et al., 2013). Moreover, the non-negligible influence of WSOC on the snow albedo

9 and radiative effect indicated that it could not only accelerate snow melting, but change the periods and

10 mass of water and carbon exchange between snowpack and underlying soils or vegetation (Meyer and

11 Wania, 2008), but also potentially affect regional climate through changing the surface radiative balance

12 (Beres et al., 2020).

1 *Data availability*. Data presented and used throughout this study can be accessed through the

2 following data repository: https://doi.org/10.5281/zenodo.6541956.

3 *Supplement*. The supplement related to this article is available online at:

4 *Author contributions*. XN and WP designed the study and wrote the first draft with contributions

5 from all coauthors. XN designed and conducted the lab experiments with the assistance of YZ and

6 HW. XN processed the data with the assistance of DW and TS. XN, WP, YC, YX, TS designed

7 and conducted the field campaign. XW supervised this study. All co-authors commented on the

8 paper and improved it.

9 *Competing interests*. The authors declare that they have no conflict of interest.

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