Response to Dr. Jacobi

We are very grateful for reviewer's constructive comments, which have helped us improve the paper quality substantially. We have addressed all comments carefully as detailed below in point-by-point responses. All referenced lines of the text are related to the revised manuscript with changes in tracking mode. Our responses start with "R:".

The authors present results from detailed measurements of the absorption determined in a number of melted snow samples collected in northwest China. The samples are grouped into different categories like rural, urban, and influenced by soil. The authors further analyzed the chemical composition of the snow samples with advanced analytical techniques to derive information on the water-soluble organic fraction at the bulk as well molecular level. This study generates a wealth of information on the composition of the snowpack in this region. Unfortunately, the number of analyzed samples and the characterization of the input sources for the different categories of organic compounds remain to limited to derive more general conclusions on the snow composition under different conditions. In a second part, the authors examined the impact of the absorption related to the organic compounds on the snow albedo and compared it to the impact of the also measured black carbon (BC). It appears that under specific circumstances the contribution of the organic compounds to the instantaneous radiative forcing can be non-negligible compared to BC. This is an important finding, especially for a region that has not received yet much attention in the literature. In summary, this manuscript describes an important exploratory study for a comprehensive chemical characterization of organics in the snow and how their impact on the absorption and the snow albedo can be analyzed. Below I have listed a number of comments that the authors may want to consider before the publication of the manuscript.

Major comments:

1. In ch. 2.1 it is mentioned that snow samples were taken in 5 cm intervals. However, no further information on these samples is later found in the manuscript. Are these

samples analyzed separately or are they mixed before or after melting? If they are analyzed separately how do the absorption or chemical profiles look like? Moreover, the parameters snow depths, snow density, and snow temperature are also mentioned. Snow depths and density are listed for each snow pit in table S3, but only an average value for the density can be found. It would be useful to provide all data for all collected samples.

R: Sorry for confusing statement. In this campaign, only surface snow, and for some sites, subsurface snow samples were collected. The sentence in line 164 has been revised as "28 surface and 8 subsurface snow samples were collected from 28 sites in Xinjiang, northwestern China during a road trip in January 2018."

The description in lines 199-205 has been revised as "The snow samples were collected in sterile plastic bags (Whirl-Pak, Nasco, WI, USA) using clean, stainless steel utensils and by scooping ~ 3 L of snow on top 5 cm at each site, resulting in ~600 mL volume of melted snow water. For several sites with snowpack deeper than 10 cm, subsurface snow (~5-10 cm) was also collected. Snow depths, snow density, and snow temperature were also measured for each sampled snow layer (Shi et al., 2020)."

For all optical and chemical measurements, 28 surface samples were analyzed. The relevant statement has been added in line 206 as "28 surface samples were analyzed by the following analytical techniques."

The values reported in Table S5 in the revised SI file are for surface snow only. A homogenous snowpack assumption was applied in snow albedo modeling, for both snow physical properties and pollutants concentrations. The statement has been added in lines 342-345 as "The input values of parameters for the SNICAR model, which are those for surface snow, are summarized in Table S5. For simplicity, a homogenous snowpack assumption was applied for both snow physical properties and pollutants concentrations."

During 2018 Xinjiang campaign, the snow depths of most sites (20/28) were lower than 10 cm, hence sampling of 5 cm is a good representative for most of the snowpack. Moreover, surface layer is the most important for optical properties of snowpack. As reported by Zatko et al. (2016a), the photic zones are 4–7 cm for snowpack from the

Uintah Basin, Utah, USA, which are much shallower compared to snow in polar regions (Zatko et al., 2016b). Due to the similar geographical conditions (latitude, sampling time, etc.) of Xinjiang and Uintah Basin along with higher snow pollutants levels in Xinjiang, the shallow photic zones for our samples can be expected. Overall, we believe that the assumption of homogenous snowpack is practical in this study.

2. Ch. 3.2: The authors compare the measured absorption with absorptions obtained in other studies described in the literature. However, the snowpack properties and further conditions during the sampling in the different studies are not sufficiently described. For example, a thin or patchy snowpack is susceptible to the input of absorbing compounds by local sources. Apparently, this was the case for some of the here reported samples. What were the conditions during the sampling for the other cited studies? For example, the samples described by Voisin et al., 2012 were collected well before the melting period with a complete snow cover, while the samples in the study by Zhang et al., 2020 were collected in April and May. Were at this time the snow and sea ice cover still intact? Depending on the size of the glaciers the local impact should be much reduced for the samples examined by Yan et al., 2016. These effects should be considered for the comparison of the results of the different studies.

R: There are two separated campaigns in Zhang et al. (2020). One was conducted at Barrow, Alaska from April to May 2017 (snow on sea ice), the other one was across Alaska in April 2017 (continental surface snow). There is no significant difference on their MAC_{365} values (0.32 vs. 0.37 m^2 g^{-1} for snow on sea ice and continental snow, respectively). The authors do not provide detail information for their sampling sites. To eliminate possible concern for the impact of snow metamorphism processes on MAC values, we have changed 0.32 m^2 g^{-1} (April to May) to 0.37 m^2 g^{-1} (April) (line 432) for discussion in our study, since snow is typically well covered at Alaska in April (Cox et al., 2017), which was also a common sampling time for earlier snow-related projects in Alaska (Beine et al., 2012;France et al., 2012). In Zhang et al. (2019), they found that the MAC values of snow WSOC increased significantly during melting season.

Relevant discussion has been added into lines 433-434. For Yan et al. (2016), they proposed that the high MAC_{365} (1.3-1.4 m^2 g^{-1}) of their glacier snow samples was attributed to dust source, which has been mentioned in line 438.

3. Ch. 3.2: Although HULIS2 is introduced as potentially stemming from marine sources, this seems not to be tested by the authors, who attribute this fraction to anthropogenic sources. Did the authors check any correlation with the sea salt components that were also measured in the samples?

R: Sorry for the misleading. Because there are very limited literature sources on snow EEM measurement, we briefly summarized the EEM application and fluorophore identification from previously published works in lines 477-487. While HULIS-2 shows similar peak positions to the marine sourced fluorophores, they do not necessarily indicate the same sources due to large differences of the physicochemical and geochemical processes (Chen et al., 2016;Duarte et al., 2007). As suggested, we did correlation analysis between sea salt ions, i.e., sodium and magnesium ions, and HULIS-2, but there are no significant correlations ($r = 0.09$ and 0.01, $p>0.05$, for sodium and magnesium ions, respectively). Xinjiang is at the center of Eurasia and thousands miles away from oceans, hence marine organics are not considered as major sources of WSOC in this study. We have added clarification in lines 487-491 as:

"Of note, the fluorophores detected in our samples show the similar peak positions compared to the previous reports for aerosol or aquatic environments, but they do not necessarily have the same sources due to large differences of the physicochemical and geochemical processes (Chen et al., 2016b; Duarte et al., 2007)."

4. L. 426: "These results provide a useful framework for representing snow BrC optical properties in climate models." This statement should be explored further or deleted.

R: This sentence has been revised in lines 474-476 as:

"These results provide a useful dataset of snow BrC light-absorbing properties which may inform climate models."

5. Fig. 4, 6, and 7: Based on a simplistic hypothesis, one could expect that the imprint determined in the soil-influenced samples would also be reproduced in the urban and rural samples with additional compounds stemming from further, potentially anthropogenic sources. While this could be deduced from the patterns shown in Figures 4, 6, and 7 for the urban samples, this is apparently not the case for the rural samples. Do the authors have any explanation why the soil-induced pattern is not present in the rural samples? In my opinion this is a topic that should be explored in the manuscript.

R: Compared to urban (U) and soil-influenced (S) samples, less allochthonous input (aerosol and local soil depositions) of snow WSOC is expected in remote/rural (R) samples, because R samples were collected from relatively remote regions with fewer anthropogenic activities and higher snow cover rate. As a result, R samples present distinctive chemical characteristics. Figures 4b and 5a show that compounds with molecular weight of 250-300 Da are most abundant in the representative R sample, which is significantly different from U and S samples. As we discuss in Sec. 3.3.2 (lines 677-687), R samples have the highest fraction of aliphatic/proteins-like species among four groups (mean: 57%±4%, Table 3 and Fig. 6). These species are commonly regarded as a marker of biological-related sources, such as microbial activities and primary bioaerosols (e.g., plant debris, fungal spores, and pollen). The high contribution of aliphatic/proteins-like species in R samples is consistent with the results of our EEM-PARAFAC analysis, which showed that PRLIS fluorophore is the most abundant. In Fig. 7, the substantially fewer unique molecules in DBE vs. C+N plots for representative R sample indicate relatively low chemical complexity of its WSOC component. Overall, according to Figs. 4-7, the chemical composition of snow WSOC from R samples is quite different from that of the other groups of samples.

Minor comments:

6. Fig. 1a: The photographs are not convincing. For example, a difference between “Grassland” and “Dessert” is not obvious to me and it is unclear how these sites are distinguished. The other two photographs do not contribute further information for

the sites.

R: While we agree that indeed no contrasting information can be obtained from the photographs, they do provide visual impression of the snow coverage in the area of study. Therefore, we opted to keep photographs as a part the artwork for Fig 1.

7. L. 213: "An ultrapure water (18.2 MΩ cm) was used for.." Not complete.

R: Revised as "An ultrapure water (18.2 MΩ cm, Milli-Q purification system, Millipore, Bedford, MA, USA)" (line 236).

8. L. 254: Please rephrase "…extracts were blown down to 200 µL…" and also "… and blew them down by pure N2." In text S1). For non-chemist this could be difficult to understand.

R: The sentence in main text has been rephrased as "Finally, the reconstituted extracts were further concentrated to 200 µL prior to HPLC analysis" (line 280). The text of S1 has also been rephrased as "Finally, we combined eluents 1 and 2 as analytes and concentrated them by pure N_2".

9. L. 312: "**field**-measured"?

R: Sorry for the ambiguity. This sentence has been revised as "The effective snow grain size was retrieved from the spectral albedo measured in the field" (line 340).

10. L. 331: "the broadband albedo (α) of each scenario **needs** to be…"

R: Corrected in line 362.

11. L. 362: "It follows that in addition to the snow and glaciers from polar or alpine regions, the seasonal snow in Northern Xinjiang is also an important organic carbon source for the covered ecosystems." The meaning of this phrase is unclear.

R: This sentence has been revised in lines 396-402 as:

"It has been reported that glaciers and ice sheets from polar or alpine regions store a large amount of WSOC and discharge it to their downstream terrestrial ecosystems

(Hood et al., 2009, 2015; Singer et al., 2012). Comparable or even higher concentrations of WSOC in our samples indicate that the seasonal snow in Northern Xinjiang is also an important organic carbon source for the terrestrial ecosystems during spring meltdown."

12. L. 366: "the mass contributions of sulfate ions at U sites (Table S1, mean: 33%±7%), which is a commonly-used marker for fossil fuel burning…" This actually applies to non-sea-salt-sulfate. By looking at the sodium fractions, the sea salt sulfate appears to be limited, but it would be useful to calculate the non-sea-salt-sulfate in this context.

R: Based on our analysis results, it is unlikely that marine sources need to be considered in Xinjiang, an inland area with thousands miles away from ocean. Specifically, calcium ion concentrations in our samples were higher than that of sodium ion, leading to much larger mass ratios of calcium to sodium (mean: 7.1±5.2) than that in seawater (0.038) (Pio et al., 2007), hence the sodium ions from marine sources in our samples are at very trace levels.

13. L. 378: "… therefore, pollutants had been potentially accumulated…" So far in the manuscript, the snow was characterized in terms of WSOC. Why are the authors are here referring to pollutants?

R: Sorry for the ambiguity, "pollutants" has been revised as "WSOC" in line 418.

14. Fig. 9: I'm not convinced that this figure is needed since the strong impact of BrC on the absorption at wavelengths below 450 nm is already obvious in Fig. 8.

R: While indeed impact of BrC on the snow surface albedo at <450nm is seen in Fig 8, its contribution in comparison to other light-absorbing components is better illustrated in Fig 9 which we believe contributes to clarity of the paper. Therefore, we opted to keep Fig 9 in the manuscript itself.

15. L. 848: “This study presents a comprehensive overview of WSOC and its BrC properties in seasonal snow of northwestern China…” I am not convinced that the limited number of snow samples can constitute a comprehensive overview. Such a characterization is also contradictory to the fact that the sample from site 120 has unique absorbing and chemical features that do not fit into the patterns found at the other sites. A more cautious statement is preferable.

R: Thanks for the suggestion. This sentence in lines 922-925 has been revised as: “This study presents a comprehensively chemical characterization of WSOC and its BrC properties in collected seasonal snow samples from Northern Xinjiang, northwestern China, which helps to better understand their characteristics, sources, and climate effects.”

16. Text S1: According to the presented data the recovered compounds account for less than 80% of the initial absorption (loss of 16% during the charging of the cartridge, another 6% loss during the two-step elution). It would be useful if the authors can explore what the potential impact of this missing fraction could be on the results and on the conclusions.

R: Compared to filter-collected aerosol samples, we cannot concentrate analytes by increasing the area of clipped filter or decreasing the volume of solvents for snow samples. Therefore, the main objective of SPE is to concentrate WSOC in snow water and derive a representative mixture of it. Only in this way, snow WSOC can be detectable for both PDA detector and HRMS. The secondary objective of SPE is desalting because salts with high concentration block the inlet of HPLC-HRMS system and decrease the ionization efficiency of ESI.

The strongly hydrophilic organics such as low molecular weight organic acids and sugars (e.g., oxalic acid, xylose, etc.) are expected to pass through the SPE cartridges, which have little light absorption (Lin et al., 2010a,b). Even if we inject them into HPLC, they probably cannot be resolved by C18 column due to very high polarity. Compounds bearing aromatic rings and multiple polar functional groups are expected to be retained, which may show strong light-absorbing properties and they are our target materials. In addition, as shown in the table below, the efficiencies of SPE (determined by UV-Vis absorption at specific wavelength) are generally less than 80% (~40% - 80%) in the literatures. Overall, the efficiency of SPE in this study is excellent and the extracted fraction is a good representative for snow WSOC.

Cartridge type	Analytical methods	Recovery (%)	References
C-18	Abs_{280}	64	Varga et al. (2001)
XAD-8	Abs_{250}	72-79	Duarte and Duarte (2005)
C-18	Abs_{250}	41-73	Fan et al. (2012)
C-18	Abs_{250}	64-73	Fan et al. (2018)
HLB	Abs_{254}	54-67	Zou et al. (2020)

17. Table S1: Like for BC and WSOC the measured concentrations of the soluble ions should be given, not only the mass fraction of the total ion mass for each specie. A further table with all measured concentrations would be useful. Moreover, all numbers should be reduced to their significant digits (also throughout the manuscript), i.e. instead of “4.53±3.06” it should be “5±3”. The mass fraction for the single site 120 should accordingly be reduced, i.e. instead of “3.98” use “4” for sodium.

R: Values of all measurements has been provided as Table S3 in the revised SI file. All numbers in the manuscript have been adjusted to appropriate decimals.

18. Fig. S4: Any unit for the “sum of squared error”?

R: Sum of squared error is unitless because it is calculated for EEM maps normalized to their total signals (Murphy et al., 2013).

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Response to referee#2

We are grateful for the reviewer's insightful comments, which are helpful and valuable for improving our manuscript. We have addressed all of the comments carefully as detailed below in our point-by-point responses. All referenced lines of the text are related to the revised manuscript with changes in tracking mode. Our responses start with "R:".

This manuscript by Zhou et al. describes the investigation for molecular composition, optical properties, and radiative effects of water-soluble organic carbon in snowpack samples from Northern Xinjiang. The results indicated that they are varied based on the geographic differences and proximity of emission sources. For example, the WSOC in S sites were characterized with high MAC_{365} values and high oxygenation and saturation levels, whereas more reduced S-containing species with high degree of unsaturation and aromaticity were identified in U samples. This study also demonstrating the important influences of WSOC on the snow energy budget. In all, the experiments were designed and conducted with cautions, and the literary presentation is excellent. I recommend publication in ACP after the authors address the following comments.

Comments

1. L 154-155: The classification of these sampling site should be also listed in a new Table or embedded in Table ***.

R: The classification scheme of sampling sites has been added as Table S1 in the revised SI file.

2. Line 165-167: In this study, the sampling sites were mainly classified based on geographical location and proximity. Why the site 120 was not be assigned into one group? Only one sample 120 is very difficult to indicate its environmental application.

R: According to the geographical location and proximity of emission sources for site

120, it is a rural/remote site. However, its chemical composition and optical properties are very different compared to R and all other samples of our study. Specifically, it has very high WSOC concentration (7069 ng g^{-1} vs. 885 ng g^{-1} of R group average, Table S2), MAC_{365} (0.95 m^2 g^{-1} vs. 0.38 ng g^{-1} of R group average, Table S2), unique UV-vis features (strong spectral feature at ~340 nm, Fig. 2) and EEM map (peak at ex/em: ~320/450 nm, Fig. S5), high contribution of lignins/CRAMs-like species (75% vs. 50% of R group average, Table 3), and so on. All these metrics indicate that site 120 does not belong to any other groups.

Site 120 is a surprise for us. Yes, it is hard to draw a general conclusion through only one sample, but at least, it shows the potential significance of plant-derived organics to optical properties, chemical composition of snow WSOC, and energy budget of snowpack.

3. Lines 362-364: Please rewritten this sentence.

R: This sentence has been rephased in lines 396-402 as:

"It has been reported that glaciers and ice sheets from polar or alpine regions store a large amount of WSOC and discharge it to their downstream terrestrial ecosystems (Hood et al., 2009, 2015; Singer et al., 2012). Comparable or even higher concentrations of WSOC in our samples indicate that the seasonal snow in Northern Xinjiang is also an important organic carbon source for the terrestrial ecosystems during spring meltdown."

4. Lines 351-364: in this paragraph, the concentrations of WSOC in U and S samples were comparably discussed based on many previous studies. Why the WSOC results in R sites were ignored?

R: The reason why we omitted the discussion of WSOC concentrations for R samples is that their values are relatively low and not much higher than the literature reports of high-altitude or high-latitude sites. Hence, we just highlighted the U and S samples with abundant WSOC in this paragraph. We did include discussion on R samples in the next paragraph (lines 413-419). Besides, several quantitative values from literature have been added for better comparison in lines 413-416:

"Although WSOC concentrations in R samples were relatively low (885±328 ng g^{-1}), they were still higher than most of the values from high-altitude or high-latitude regions of previous studies (~40 – 700 ng g^{-1} as mentioned in the last paragraph)."

5. Lines 374-376: how about the data in previous studies?

R: Please see response to comment #4.

6. Lines 428-437: One concern is the data processing of EEM: In fact, the three fluorescent components are not relevant with humic-like and protein-like substances that derived from water (phytoplankton)/terrestrail soil, but they should be resided on the same peak positions. Author should give some explanation like this way using references, otherwise, some readers can mistake to have similarly or dissimilarly.

R: We really appreciate the suggestion. Because there are very limited literature reports on snow EEM measurement, to give readers a general idea about how to explain EEM results, we briefly summarized the fluorophores identification from other scientific community in this paragraph. Clarification has been added in lines 487-491:

"Of note, the fluorophores detected in our samples show the similar peak positions compared to the previous reports for aerosol or aquatic environments, but they do not necessarily have the same sources due to large differences of the physicochemical and geochemical processes (Chen et al., 2016b; Duarte et al., 2007)."

7. Lines 452-453: the statistical correlation between HULIS-1 and nitrates don't necessarily mean it is scientific reasonable. Please add some references to support that.

R: More discussion has been added in lines 510-512 as:

"For instance, Yang et al. (2018) found that HONO formation is significantly enhanced, in the presence of humic acid, from nitrate photolysis."

8. Figure S5: how can you obtain this Figure.

R: It is a direct output by *eemview* function of drEEM toolbox (version 0.2.0, http://models.life.ku.dk/drEEM).

9. Lines 546-548: The relative content of CHON+ cannot simply indicated they from aerosol depositions rather than from autochthonous sources.

R: Thanks for the suggestion. Our logic here is that the CHON fractions of our samples are comparable with the fresh snow samples in Feng et al. (2018, 2020), in which the major source of WSOC is aerosol wet/dry deposition. However, during snow metamorphism, the CHON fraction increased to ~40% (Feng et al., 2018, 2020), which is mainly attributed to microbial activities in snow. Based on the discussion above, we conclude that WSOC in our samples is more likely from aerosol depositions rather than autochthonous sources. The sentences in lines 602-610 have been revised as:

"The fractions of CHON+ species in our samples were significantly lower than those of the aged firn/ice samples from the TP (~40%) (Feng et al., 2016, 2018, 2020; Spencer et al., 2014) which is mainly attributed to less microbial activities in our samples; but were comparable with those of fresh snow (Feng et al., 2018, 2020) in which the major source of WSOC is aerosol wet/dry deposition. These results indicate that WSOC from the snowpack in northern Xinjiang was more likely from atmospheric aerosol depositions rather than from autochthonous sources."

10. Lines 576-577: This is only the data in ESI+ mode. As shown in Table 2, R samples showed higher DBEw and AIw values. Why the unsaturation and aromaticity of WSOC detected at ESI+ and ESI- are different for these samples? Please give more discussion.

R: The sentence in line 638-639 has been revised as "The U samples showed higher DBE_w and AI_w values than the other groups of samples mainly due to high fractions of S-containing compounds in ESI+ mode (Table 1)."

In addition, the discussion on different molecular characteristics of compounds detected in ESI+/– modes has been added in lines 648-654:

"Additionally, the molecular characteristics of formulas detected in ESI− and ESI+ are different, e.g., higher average DBE_w and AI_w values of R samples for ESI− data (Tables 1 and 2). This results from the differences of ionization mechanisms between positive and negative modes. ESI+ is sensitive to protonatable compounds with basic functional groups, while acidic species are easily deprotonated and detected in ESI− mode (Cech and Enke, 2001). Therefore, using both positive and negative ESI modes provide a more complete molecular characterization of WSOC."

11. Lines 600-602: add references to support that.

R: Added in line 672.

12. Lines 623-627: I cannot understand this sentence.

R: Revised lines 694-699 as "In addition, the S samples showed higher contributions of unsaturated hydrocarbons (mean: 19%±14%) than the R samples and the site 120 sample (mean: 11%±2% and 10%, respectively). This result is mainly due to the CHOS+ compounds detected in the site 104 sample from R group (Fig. 6g), indicating some influence by anthropogenic pollution at this site."

13. Fig 7: In many previous studies, plots of DBE versus C atomic number were shown, why plots of DBE versus C+N atomic number rather than C atomic number were shown in this study?

R: According to Eq. (3) in the main text: DBE = C-H/2+N/2+1, in which N atom is also involved into DBE calculation. Accordingly, DBE/C values for CHON compounds would be higher than those species with the same C and H atom numbers but without N. The references lines in the DBE vs. C+N space are derived from hydrocarbons. Hence, some CHON compounds can above the fullerene-like carbon limit. To address this problem, C+N was employed for x-axis in Fig. 7.

14. Lines 724-728: The total rather than unique "BrC" molecules should be used to investigated for their correlation with their bulk MAC values.

R: We really appreciate this constructive suggestion. We realized that relating numbers of "BrC" molecules with bulk MAC values may be misleading, because the total absorption of WSOC might be determined by small amount of strong light-absorbing molecules (Laskin et al., 2014). We have deleted these sentences in lines 797-801.

References

Laskin, J., Laskin, A., Nizkorodov, S. A., Roach, P., Eckert, P., Gilles, M. K., Wang, B., Lee, H. J., and Hu, Q.: Molecular selectivity of brown carbon chromophores, Environ. Sci. Technol. , 48, 12047-12055, 10.1021/es503432r, 2014.

Letter to the editor

During the revision, we found an erroneous assignment of MS peaks detected at ‘site 120’ sample in ESI– mode. This assignment was corrected, as highlighted by the red boxes in Fig. 4 of the tacking mode file. Accordingly, the molecular metrics in Table 2, and Figs. 4, 6, and 7 have been edited (only one bar/dot in each figure). The discussion related to Table 2 has been also revised in lines 641-647. All these changes are included in the tracking mode revised manuscript. We note that these corrections do not change any discussion points or conclusions of our manuscript.