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.