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The Cryosphere
Discussions

Interactive comment on "The optical characteristics and sources of chromophoric dissolved organic matter (CDOM) in seasonal snow of northwestern China" *by* Yue Zhou et al.

Anonymous Referee #2

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Review of "The optical characteristics and sources of chromophoric dissolved organic matter (CDOM) in seasonal snow of northwestern China" by Zhou et al, submitted to The Cryosphere

General comments

This work describes the results of a field campaign on surface snow chemical properties, lead in northwestern China. More specifically, it investigates the colored dissolved organic matter (CDOM) in seasonal snow, trying to evaluate its different components, their sources, and overall importance for light absorption by snow, which is a very important topic, linked to the climate impact of snow. The subject treated is thus highly

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relevant for The Cryosphere and is worth publishing, once the authors take care of the following remarks.

My major issue with this work is related to clarity. Although generally well written, I truly think the authors should make a distinct effort on two aspects: - Have a clearer presentation of the PARAFAC method. Although as stated by the authors, it has become very mainstream in the aquatic chemistry community, and is starting to be used in the aerosol community, it is still a novelty for most readers of The Cryosphere. It would be good, in this case, to have a clear reminder of what the PARAFAC methods gives (what are the Components, the Fmax, . . .). If the authors are somewhat familiar with the PMF method, they might even want to draw a parallel, which might (arguably) help - Maybe draw a clearer separation between actual results, and their interpretation in terms of sources and comparison with previous studies. This could be done by adding a "discussion" section. In particular, this might help clarifying the case on sources. In its current form, the paper discusses sources through the analyses of PARAFAC components, and then though the analyses of backtrajectories and other data such as the ion data. I feel that the case of the authors on sources would much stronger if raw results were presented first (PARAFAC components, clusters, ion ratios, maybe back-trajectories) and then discussed together: this would help synthesis, and avoid losing the reader between two different discussions on the same topics.

Specific comments:

P7 line 14: there were studies on EEM application to aerosols before the recent papers cited here. Please refer to the appropriate literature (probably not exhaustive): - Duarte, R. M. B. ., Pio, C. A. et Duarte, A. C.: Synchronous scan and excitation-emission matrix fluorescence spectroscopy of water-soluble organic compounds in atmospheric aerosols, Journal of Atmospheric Chemistry, 48(2), 157–171, 2004. - Lee, H. J. (Julie), Laskin, A., Laskin, J. et Nizkorodov, S. A.: Excitation–Emission Spectra and Fluorescence Quantum Yields for Fresh and Aged Biogenic Secondary Organic Aerosols, Environ. Sci. Technol., 47(11), 5763âĂŚ5770, doi:10.1021/es400644c,

2013. - Mladenov, N., Alados-Arboledas, L., Olmo, F. J., Lyamani, H., Delgado, A., Molina, A. et Reche, I.: Applications of optical spectroscopy and stable isotope analyses to organic aerosol source discrimination in an urban area, Atmospheric Environment, 45(11), 1960âĂŚ1969, doi:10.1016/j.atmosenv.2011.01.029, 2011.

P8 line 18: “grouping scheme presented by Pu et al. (2017)”: Pu et al actually just refer to “geographical distribution” as a grouping scheme. Reading further in the article, there is a logic, in terms of north or south of a mountain range, on this or that side of strong potential human sources, . . . I suggest this logic should be somewhat detailed here, rather than reporting to a reference where it is actually not clearly presented

P10 line 9-10: “In addition, the emission wavelengths longer than 650 nm were removed to eliminate the uncertainty of measurement”. It is not clear what uncertainty is eliminated here. Please be more specific.

P10 line 10-15: as in any statistical factor analysis (PARAFAC, PMF, . . .) the choice of factor number is quite critical, and thus must be very carefully argumented. Here, the choice of 3 components is based on residual error analysis. Yet, although going from 2 to 3 decrases strongly this error, there is still (fig S1) a large error around 270nm which desapears when going from 4 to 5 factors. The authors “confirm” the 3 factor analysis with some splitting method, but they do not reject the 4 or 5 factor analysis with this method. To me, it seems at this point, the choice of 3 factors is largely arbitrary.

P10: I may have missed it, but did the authors mention the number of samples used in the PARAFAC analysis? Is that number sufficient for such a statistical method? There are quality guidelines for PMF studies from filters and offline tracers analysis (Belis et al, 2014), I would expect similar guidelines to exist for PARAFAC, as these methods are mathematically very close (if not equivalent)

P11 line 17-19: the reason put forward by the authors for choosing one fitting method rather than the other seems statistically weak to me. Of course, as any point where the choice would actually matter is anyway rejected in advance, it is of minor importance.

Yet, could the authors be more specific here ?

P11 line 17-25 : confidence on fits ? this translates in uncertainties in the reported slopes and AAE, which might impact interpretations. So it is of some importance !

P12 line 20: nss-K is reportedly calculated after Tao et al 2016 following nss-K = K - 0.159 Mg. Tao et al 2016 actually claim they used Cheng et al, 2000 definition of nss-K, reported as nss-K = K - 0.037 Na, which they claim they took from Hitchcock et al, 1980. I would suggest being more precise on the calculation really made, and its origin. In any case, I also have some reservations on this approach, as it was originally used for coastal sites (North Carolina for Hichcock et al, 1980 ; Hong Kong for Cheng et al, 2000). In more continental areas, with significant input of terrestrial dust, there might be a sizable portion of either Mg2+ and/or Na+ coming from dust, which would distort the relation used. An example of such distortion and the way to analyse it is presented in Pio et al, 2007. I suggest to take these points into account.

P13 line 7-8 : how was the choice of 4 clusters decided to be relevant ?

Fig 5: replace “excitaition” with “excitation” ; it is not quite clear to me what “excitation (emission) loading” is. I suspect it is the sum over emission (excitation) of the EEM matrix of each component. Am I right? maybe it should be precised either here in the caption, or somewhere in the text (or both).

P19 line 4-8: It is not clear to a non-PARAFAC aficionado what Fmax is. I feel that it is the fraction of the observed total fluorescence that is explained by a given component, so that it should be somewhat proportional to the concentration of this component in the mixture that the sample is made of. It would be really helpful to clarify this point.

P21 line 9: the picture presented by the authors looks much like lichens to me, more than algae. I also thought that algae only lived in aquatic media (including snow), except when associated with fungi in lichens.

P22 line 18: I feel that figure 9 is not very informative as such

P23 line 2-5: Figure S6 should be considered as a replacement for figure 10: it is more informative, while not being really more clutered. Maybe it could be made even better by reducing slightly the font size of the red equations and drawing dashed red lines, thus using size and line type and color to make stress which correlation is important and which one is rejected.

P25 line 20: see my previous comment on nss-K+. how is the authors discussion here sensitive to the objection raised in that previous comment?

P26: I have a hard time evaluating whether the backtrajectory analysis presented her is relevant. Obviously, fires location from MODIS can be active or not when a backtrajectory passes over it. Here, it seems that any backtrajectory can overpass any fire location, and be taken into account in the analysis, even if the backtrajectory overpasses on day n, and the fire was only active from day n+3 to day n+10. This seems to weaken a lot the analysis presented here.

Belis, C. A., Favez, O., Harrison, R. M., Larsen, B. R., Amato, F., El Haddad, I., Hopke, P. K., Nava, S., Paatero, P., Prévôt, A., Quass, U., et al.: European guide on air pollution source apportionment with receptor models., Publications Office, Luxembourg. Available from: http://dx.publications.europa.eu/10.2788/9307, 2014.

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Hitchcock, D. R., Spiller, L. L. et Wilson, W. E.: Sulfuric acid aerosols and HCl release in coastal atmospheres: Evidence of rapid formation of sulfuric acid particulates, Atmospheric Environment (1967), 14(2), 165âĂŚ182, doi:10.1016/0004-6981(80)90275-9, 1980.

Pio, C. A., Legrand, M., Oliveira, T., Afonso, J., Santos, C., Caseiro, A., Fialho, P.,

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Barata, F., Puxbaum, H., Sanchez-Ochoa, A., Kasper-Giebl, A., et al.: Climatology of aerosol composition (organic versus inorganic) at nonurban sites on a west-east transect across Europe, J. Geophys. Res., 112(D23), doi:10.1029/2006JD008038, 2007.

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