Responses to the comments (bgd-12-C1351-2015):

Thank you very much for recommending the reference "Mostofa KMG, Liu CQ, Yoshioka T, Vione D, Zhang YL, Sakugawa H (2013) Fluorescent dissolved organic matter in natural waters. In: MostofaKMG, Yoshioka T, Mottaleb A, Vione D (Eds), Photobiogeochemistry of Organic Matter: Principles and Practices in Water Environments, Springer, New York, Chapter 6, pp429-559." The authors have read this reference carefully.

1) "fluorescence components" should be replaced by "fluorescent components" throughout the manuscript.

Response 1: Thank you very much for pointing out the mistakes.

Your kind suggestion has been adopted in the revised manuscript; "fluorescence components" were replaced by "fluorescent components" throughout the manuscript.

2) Author expression of the fluorescent components are needed to change throughout the manuscript. For example, in the abstract: "Two humic-like peaks C1 (Ex/Em= 230, 300/425 nm) and C2 (Ex/Em= 255, 350/460 nm)" here, two humic-like components, not peaks. Similarly, "and two protein-like B (Ex/Em= 220, 275/320 nm) and T (Ex/Em = 225, 290/360 nm) peaks", here also the same mistakes and also author should separate them easily, not saying two protein-like components: first one should be tyrosine and second one should be "Tryptophan" based on the excitation emission fluorescence peaks. That should be needed to change throughout the manuscript.

Response 2: Thank you very much for pointing out the wrong expression of the fluorescent components.

The contents "Two humic-like peaks C1 (Ex/Em= 230, 300/425 nm) and C2 (Ex/Em= 255, 350/460 nm)" should be replaced by "Two humic-like components C1(Ex/Em= 230, 300/425 nm) and C2 (Ex/Em= 255, 350/460 nm)".

The contents "and two protein-like B (Ex/Em= 220, 275/320 nm) and T (Ex/Em = 225, 290/360 nm) peaks", should be replaced by "and the protein-like components

(C3 and C4): tyrosine-like B (Ex/Em= 220, 275/320 nm) and tryptophan-like T (Ex/Em = 225, 290/360 nm).” throughout the manuscript. Your kind correction has been adopted in the revised manuscript.

3) “The humic-like component is a complex mixture of aromatic and aliphatic compounds fulvic acids and humic acids, which were originated from terrestrial materials or algal decomposition in the waters. While the two protein-like components consist of two dissolved amino acids, i.e., tryptophan and tyrosine...”. Wording and discussion are completely wrong in these sentences. Authors need more study in that issue to distinguish the terrestrial humic substances which are composed of humic and fulvic acids. But aquatic humic-like substances are of algal or phytoplankton origin. Authors need clarify in that regards. Second issue, I have explained in my second comments. “..the two protein-like components consist of two dissolved amino acid..” it is entirely wrong. These issues needed them clarify well.

Response 3: Thank you very much for pointing out the wrong expression. Your kind correction was accommodated in the revised manuscript.

The contents “The humic-like component is a complex mixture of aromatic and aliphatic compounds fulvic acids and humic acids, which wereoriginated from terrestrial materials or algal decomposition in the waters. While thetwo protein-like components consist of two dissolved amino acids, i.e., tryptophan andtyrosine…” should replace by the contents “The humic components is a mixture of aromatic and aliphatic compounds from terrestrial substances composed of humic and fulvic acids, and aquatic humic-like substances of phytoplankton origin. While the protein-like components-tyrosine-like (B) and tryptophan-like (T), mainly consist of dissolved amino acids.

Responses to the comments (bgd-12-C3088-2015):

One key point of this paper that I missed from my earlier comments:

When I got the response from author comments, I have been checked once again the research paper. At the same time, I also read the one of the referee comment. My missing point is that author used the Raman Unit (cm-1) that can change the excitation-emission wavelengths and also at the same time the fluorescence intensity of the fluo-rescence peaks. Such effect is problematic for future understanding of the fluorescence spectra of various components. Author can read this issue in the reference paper that I have been provided before. It has been elaborately discussed how the Raman Unit affects the peak wavelengths and also the intensity. I strongly suggest using the arbitrary unit (a.u.) or standard Quinine sulfate unit (QSU) instead of Raman Unit (cm-1) and then rewrite the sections which are needed throughout the manuscript.

Response: Thank you very much for recommending the reference "Mostofa KMG, Liu CQ, Yoshioka T, Vione D, Zhang YL, Sakugawa H (2013) Fluorescent dissolved organic matter in natural waters. In: MostofaKMG, Yoshioka T, Mottaleb A, Vione D (Eds), Photobiogeochemistry of Organic Matter: Principles and Practices in Water Environments, Springer, New York, Chapter 6, pp472-475." The authors have read this reference carefully.

To compare the fluorescence data with the results from different fluorescence spectrometers, the fluorescence intensity calibration have been used, which has been expressed by three methods in the published reports:

1) Quinine sulfate (QS) calibration method is usually used in fluorescence spectroscopy. But different QS concentrations and solvents have established different scaling units, such as millifluorescence (mFI), the fluorescence unit (flu) and the quinine sulfate unit (QSU). This method requires separate measurement daily for each instrument and makes it difficult to produce a unified fluorescence standard as an absolute calibration (Determann et al., 1994. Fluorescent matter in the eastern Atlantic Ocean. Part 1: methods of measurement and near-surface distribution; Determann et

al., 1996. Fluorescent matter in the eastern Atlantic Ocean. Part 2: vertical profiles and relation to water masses). Because a number of investigators have used QSU to calibrate the fluorescence data (Coble, 1996; Zhang et al., 2010, 2011), QS solution will be measured daily in our future study.

2) The integrated area of water Raman peak is a rapid and simple method for fluorescence intensity across different instruments expressed by Raman units (nm^{-1}), which makes this quantification without no need of fluorescence standards (Lawaetz and Stedmon, 2009). The Raman scattering of water shows a signal included in each seawater spectra with a bandwidths related to the excitation line, which have been reported by Determann et al., 1994 and 1996 (Determann et al., 1994. Fluorescent matter in the eastern Atlantic Ocean. Part 1: methods of measurement and near-surface distribution; Determann et al., 1996. Fluorescent matter in the eastern Atlantic Ocean. Part 2: vertical profiles and relation to water masses). Compared with the Quinine sulfate (QS) calibration, this method would require no separate measurement for each instrument and has been widely used in the earlier studies (Stedmon et al., 2003；Stedmon and Markager, 2005; Stedmon and Bro 2008; Jaffe´ et al., 2008. Spatial and temporal variations in DOM composition in ecosystems: The importance of long-term monitoring of optical properties; Fellman et al., 2010; Guo et al., 2011. Assessing the dynamics of chromophoric dissolved organic matter in a subtropical estuary using parallel factor analysis; Seredyn´ska-Sobecka et al., 2011. Monitoring organic loading to swimming pools by fluorescence excitation-emission matrix with parallel factor analysis (PARAFAC)).

3) The fluorescence intensity is given in arbitrary units (A.U.). This direct method makes it difficult to compare the difference across different instruments (Lawaetz and Stedmon, 2009).

Since the reference “Mostofa KMG, Liu CQ, Yoshioka T, Vione D, Zhang YL, Sakugawa H (2013) Fluorescent dissolved organic matter in natural waters. In: MostofaKMG, Yoshioka T, Mottaleb A, Vione D (Eds), Photobiogeochemistry of Organic Matter: Principles and Practices in Water Environments, Springer, New York, Chapter 6, pp472-475.” has pointed out that the RU calibration may produce unusual fluorescent components which is not shown in the original EEMs, we will use standard Quinine sulfate unit (QSU) instead of Raman Unit (nm^{-1}) in future study.

In our papers, the Raman scatter peak of water has been measured daily as fluorescence intensity calibration. This method is expressed by Raman Units (R.U.) .

Responses to the comments (bgd-12-C4015-2015) of Reviewer 2:

Overall: the language needs corrections, additional check and corrections by native speaker would be advised. Authors should more careful interpreting the components. And there are way too many methodological details in the manuscript.

Response: Thank you for your suggestions. The fourth co-author (Professor Lin Li from IUPUI, USA) has corrected the language carefully. The components were interpreted more carefully in the revised manuscript. As suggested by the reviewer, these unnecessary methodological details in the manuscript were removed. Your kind suggestion has been adopted in the revised manuscript.

Abstract The abstract should be written in more concise way. Right now it is quite confusing. Only the most important findings should be emphasized (which is not the case now). The less important findings should be removed from abstract. I do not really understand what authors mean by the sentence line 14-15?

Response: Thank you for your suggestion. The authors have rewritten the abstract and a more concise abstract was achieved in the revised manuscript. The revised manuscript is written as "The seasonal characteristics of fluorescent components in CDOM for lakes in the semi-arid region of Northeast China were examined by excitation-emission matrices fluorescence and parallel factor analysis (EEM-PARAFAC). Two humic-like (C1 and C2) and two protein-like (C3 and C4) components were identified using PARAFAC. The average fluorescence intensity of the four components differed under seasonal variation from June and August 2013 to February and April 2014. Components 1 and 2 exhibited strong linear correlation ($R^2 = 0.633$). Significantly positive linear relationships between CDOM absorption coefficients a(254) ($R^2 = 0.72$, 0.46, $p < 0.01$), a(280) ($R^2 = 0.77$, 0.47, $p < 0.01$), a(350) ($R^2 = 0.76$, 0.78, $p < 0.01$) and F_{max} for two humic-like components (C1 and C2) were exhibited, respectively. A significant relationship ($R^2 = 0.931$) was found between salinity and DOC. However, almost no obvious correlation was found between salinity and EEM-PARAFAC extracted components except for C3 (R^2 =

0.469). Results from this investigation demonstrate that the EEM-PARAFAC technique can be used to evaluate the seasonal dynamics of CDOM fluorescent components for inland waters in semi-arid regions of Northeast China; and to quantify CDOM components for other waters with similar environmental conditions. ”

Introduction: In general the introduction is too long, it should be shortened. Page 5727, line 10: reformulate “terrestrial imported substance” Too many technical details in the introduction: (page 5727, line 22-28). It is not necessary to describe it. There are repetitions. For example it is not necessary to introduce the two main components twice (page 5727, line 20 and 5728, line 17). Page 5729, line 11: I did not understand what was the actual motivation to perform this research? The novelty of this research has to be emphasized.

Response: Thank you for your suggestion. The introduction has been shortened. Your kind suggestion has been adopted in the revised manuscript.

On page 5727 in line 10, the content “terrestrial imported substance” was replaced by “terrestrially imported substance”.

On page 5727 from line 22-28 and On page 5728 from 1-2, the content was removed.

The contents on page 5729 in line 11 have been revised.

The Songnen plain is a fluvial plain with semi-arid climate, in which many fresh and brackish waters are distributed due to its geomorphology (Song et al., 2013). Dissolved organic carbon (DOC) characterisitics of these fresh and brackish waters across the Songnen Plain have been studied by Song et al. (2013); the results indicated that a huge amout of DOC were stored in these waters. In particular, brackish waters would exhibit high average DOC concentration and significantly contributed the carbon budget to inland waters (Duarte et al., 2008; Song et al., 2013; Tranvik et al., 2009). However, little studied has been made on the detailed information of DOC sources for these waters in the Songnen Plain. Therefore, it motivated us to investigate the components in CDOM for both fresh and brackish waters in the semi-arid region.

Materials and Methods

Overall too long, way too many methodological details. However I am missing the description of absorption coefficients? Which ones did authors calculate? What these absorption coefficients indicate?

Response: Thank you for pointing out the questions. The absorption coefficient a_{CDOM} was calculated from the measured optical density (OD) of the sample using Eq. :

$$a_{CDOM}(\lambda) = 2.303[OD_{S(\lambda)} - OD_{(null)}]/\gamma$$

which was explicitly described on page 5731 from line 15-20.

The absorption coefficient a_{CDOM} and the spectral slope S were calculated.

These absorption coefficients indicate the absorption intensity of CDOM in the UV and visible region of spectrum.

Why these lakes were sampled during these four times? (page 5730, line line 11) To obtain information about seasonal changes, rainfall, dry season or what was the reason behind it?

Response: On page 5730 in line 11, the lakes were sampled four times at different seasons to assess the dynamics of individual fluorescent components under seasonal variation in the Songnen Plain. For a long and cold winter, the lakes in the study region were frozen in February. The optical properties of ice-melt and the under-ice sea water have been studied in other papers (Stedmon et al., 2007; Thomas K., 1983; Uusikiv et al., 2010). After April, the ice-covered lakes were just melting into water. In order to study the differences of fluorescent components in CDOM for these lakes from ice-covered to melted-water, two seasons in February and April were chosen. With dry and windy seasons, spring and autumn are very short. The season in June is at the turn of spring and summer. In August, there is plenty of rainfall which results in flooding in summer. The CDOM in rainwater for other region has been studied by the findings of Cheng et al. (2010) (in Chinese). To study the properties of CDOM in lake-water after flooding, the samples were sampled in August 2013. Therefore, the lakes were sampled during these four times which can present obvious seasonal variation for fluorescent components.

Not necessary to explain the unit (page 5730, line 19)

Response: Thank you for your suggestion. On page 5730 in line 19, the content "…with the µScm^{-1} (micro-Siemens/centimeter) unit at room temperature (20 ± 2°C) in laboratory... " was removed in the revised manuscript.

In the section "2.3 DOC concentration measurement", the citation of the method is missing.

Response: Thank you for pointing out the question. APHA 1998 and Song et al. (2011) were cited in the method of DOC concentration measurement (APHA/AWWA/WE F.: Standard Methods for the Examination of Water and Wastewater, Washington, DC, 1998; Song, C. C., Wang, L. L., Guo, Y. D., Song, Y. Y., Yang, G. S., and Li, Y. C.: Impacts of natural wetland degradation on dissolved carbon dynamics in the Sanjiang Plain, Northeastern China, J. Hydrol., 398, 26-32, 2011).

In section "2.6 The PARAFAC modeling" it is not necessary to describe PARAFAC model, appropriate citations would be enough. This PARAFAC section was shortened significantly in the revised manuscript.

Response: Thank you for your suggestion.

On Page 5733 from line 4-22, the content was removed. Your kind suggestion has been adopted in the revised manuscript.

On page 5733 in line 4, The content "Parallel factor analysis (PARAFAC) ……" should be replaced by "PARAFAC, ……".

Section "2.7 Statistical analysis" Page 5734, line 18 and 19: correct "analyses" to "analysis" Did the authors check if the data were distributed normally? If not, then t-test can't be applied.

Response: Thank you for pointing out the error. On page 5734, in line 18 and 19, the word "analyses" was replaced by "analysis".

We are sorry for making the error. The regression and correlation analysis in SPSS was used in the paper. On page 5734 in line 19, the content "paired t test" was removed.

Results and discussion

Overall: too many methodological details, too little of actual discussion.

Response: Thank you for your suggestion. The unnecessary methodological details were removed and more discussion in the section 'Results and Discussion' was added in the revised manuscript.

"3.1 Water quality conditions" Why do authors pool all the samples together? It would be important to show the differences between fresh and saline lakes during different seasons. Page 5735, line 8 "salinity with other three seasons" should be reformulated. Repetition of word "reduced" (page 5735, line 10)

Response: For the data in the paper, there was no obvious difference between fresh and saline lakes as shown in the tables and figures. Also, these brackish and fresh water are endowed with similar geological, hydrological and climatic settings, thus we presume that similar process may control the CDOM components. To study the differences of the four fluorescent components for both fresh and saline lakes under seasonal variation from June and August 2013 to February and April 2014, we pooled the samples together. The differences of fluorescent components in CDOM between fresh and saline lakes during different seasons should be studied in further study, particularly with larger saline gradients as presented in Song et al., (2013). However, the salinity is not significant different from two groups of waters, thus all data were pooled together, and the two groups of lakes were only divided according to the study region rather than salinity.

On page 5735 in line 8, the content "salinity with other three seasons…" was replaced by "relatively constant values (around 0.40 PSU) were exhibited in the other three seasons" in the revised manuscript.

Thank you for your suggestion. On page 5735 in line 11, the content "and then reduced…" was replaced by "and the lowest value of… was recorded in February 2014".

Section "3.2 EEMs characterization of CDOM" Again too many methodological details, they should be removed.

Response: Thank you for your suggestion. The unnecessary methodological details were removed in the revised manuscript. Your kind suggestion has been adopted in the revised manuscript.

Page 5735 line 20: reformulate "While the two protein-like components consist of two dissolved amino acids, i.e., tryptophan and tyrosine that are originated from microbial..." One component is tyrosine-like only and another is tryptophan-like only, not both.

Response: Thank you for your suggestion. The content "While the two protein-like components consist of two dissolved amino acids, i.e., tryptophan and tyrosine that are originated from microbial..." was replaced by "With respect to the protein-like components, i.e., tyrosine-like and tryptophan-like substance, mainly consist of dissolved amino acids." in the revised manuscript.

It is not necessary to provide the Ex/Em of each component again (page 5735, line 24-26). The description of the components was already provided before by the authors, it is repetitive here. Page 5736, line 1-2. This sentence should be reformulated "The measured peak intensity of these fluorescence centers is dependent on the concentration of the main fluorophores dissolved in water bodies". Page 5736, line 3-15: this part belongs rather to the method section.

Response: Thank you for your suggestion. On page 5735 from line 24-26, it only show the Ex/Em of each component at approximate wavelength range based on EEMs 'peak-picking' technique. When identified by PARAFAC modeling, each fluorescent component was characterized by specific Ex/Em wavelength On page 5736 from line 16-28. We have revised the section on page 5735 from 24-26. Your kind suggestion has been adopted in the revised manuscript.

On Page 5736, line 1-2, the content "The measured peak intensity of these fluorescence centers is dependent on the concentration of the main fluorophores dissolved in water bodies" was replaced by "The measured fluorescence intensity is dependent on the concentration of dissolved fluorophores in waters" in the revised manuscript.

On page 5736 in line 3-5, the content "" To determine the appropriate number of

PARAFAC components, the split-half validation procedure was executed to verify whether the model was valid by comparing the emission and excitation loadings from each half (Stedmon and Bro, 2008)" was placed on page 5734 in line 11 in the method section.

Section 3.3 "Temporal distribution of the PARAFAC components" and **section 3.4** "CDOM vs. EEM-PARAFAC extracted components"

Overall: The general patterns and only the main findings of the results should be just emphasized. These two sections should be re-written, otherwise the reader is lost. And discussion of the results should be more extensive.

Response: Thank you for your suggestions. The general patterns and the main findings of results were emphasized. Section 3.3 and section 3.4 were re-written. More discussion of the results were added. Your kind suggestion has been adopted in the revised manuscript.

Page 5737, line 8: Why would authors pool the results from all the samples together? Again, I think it would be good to show the differences between different types of lakes, seasons or flood and no flood samples. However there are too many results, describing just patterns (increase, decrease). These two sections should be shortened and only main findings emphasized. For me as a reader, the most important results and findings are not clearly stated. Page 5738, line 9: "protein-like peaks" should be replaced "protein-like components" Page 5738, line 14: "reached to" should be replaced by "reached" Page 5739, line 1: "quantities of rainfall taking place" should be reformulated. Page 5740, line 1: what are "common sources" ?

Response: Thank you for your suggestion. We pooled the results from all the samples together in order to study the common characteristics of fluorescent components under seasonal variation. The samples at different seasons showed different characteristics in the next paper from page 5737-5738. The two sections were revised and show the important results. Your kind suggestion has been adopted in the revised manuscript.

On page 5738 in line 9, the content "protein-like peaks" was replaced by "protein-like components".

On page 5738 in line 14, the content “reached to” was replaced by “reached”.

On page 5739 in line 1, the content “quantities of rainfall taking place” was replaced by “quantities of rainfall take place”.

On page 5740 in line 1, the content “common source” refers to a common processing mechanisms leading to similar dynamics (Zhang et al., 2010, 2011).

Page 5741 line 1, 2, 5 Authors should use “components” rather than “peaks” Page 5741 line 4-6 The sentence “The lowest C2 represents only a small portion of CDOM from terrestrial import to water bodies through rainwash and soil leaching” should be reformulated Page 5741 line 6: “intensities” change to “components”

Response: Thank you for your suggestion. On Page 5741 in line 1, 2, 5, the content “peaks” was replaced by “components”.

On Page 5741 in line 4-6, the content “The lowest C2 represents only a small portion of CDOM from terrestrial import to water bodies through rainwash and soil leaching” was replaced by “At all four seasons, the fluorescent component C2 contributed less to total fluorescence than the other three.”

On Page 5741 in line 6, the content “intensities” should be replaced by “components”.

Figures: Figure 2. What is the purpose of presenting figure of this one specific sample?

Response: In Figure 2, it present examples of EEMs for one water sample from Xindianpao Lake in the western part of Jilin province at different seasons which showed that the fluorescence properties of CDOM differed under seasonal variation. For Xindianpao, the protein-like fluorescence peaks were higher than the humic-like fluorescence peaks in June 2013, whereas, the humic-like fluorescence peaks were higher than the protein-like peaks in August 2013.

Figure 5. It would be good to present seasonal variations of Fmax of each component in the separate figures. Right now there is too much information in one figure. Also it would be interesting to see if there was any correlation between rainfall and any of the

components?

Response: Thank you for your suggestion. The F_{max} of seasonal average of the components in Figure 5 was separated from the four single seasons in the revised manuscript.

The rainwater, which also contained much lower humic-like concentration, was mostly characterized by protein-like components. The correlation between rainfall and any of the components will be studied in future study.

Responses to comments (bgd-12-C2577-2015) of Reviewer 1:

Thank you very much for pointing out some major points which should be reconsidered.

- **The manuscript** contains too many methodological details. To increase the attention of the reader and provide a fluent reading the Introduction as well as the Materials and Methods should be shortened rigorously. Similarly, details about e.g. fluorescent wavelength regions or statistics that can be found in one of the tables do not need to be mentioned in the text again

Response: Thank you for your suggestion. The introduction as well as the Materials and Methods were shortened rigorously (see responses to specific comments from Page 5727 to 5734). and the fluorescent wavelength regions or statistics mentioned again in the text was skipped (see responses to specific comments).

- **As mentioned** in the introduction, an initial motivation for this manuscript was to compare fresh water and brackish water CDOM. Therefore two groups of lakes were defined (one group with low, the other with medium salt concentration). Though this might be a quite interesting investigation these two lake groups are not mentioned in the results and discussion part anymore. Were there no significant differences? If so, how could this be explained?

Response: The Songnen plain is a fluvial plain with semi-arid climate, in which many fresh and brackish waters are distributed due to its geomorphology (Song et al., 2013). Dissolved organic carbon (DOC) characterisitics of these fresh and brackish waters across the Songnen Plain have been studied by Song et al. (2013); the results indicated that a huge amout of DOC were stored in these waters. In particular, brackish waters would exhibit high average DOC concentration and significantly contributed the carbon budget to inland waters (Duarte et al., 2008; Song et al., 2013; Tranvik et al., 2009). However, little studied has been made on the detailed information of DOC sources for these waters in the Songnen Plain. Therefore, it motivated us to investigate the components in CDOM for both fresh and brackish waters in the semi-arid region.

For the data in the paper, there was no obvious difference between fresh and saline lakes as shown in the tables and figures. Also, these brackish and fresh water are endowed with similar geological, hydrological and climatic settings, thus we presume that similar process may control the CDOM components. The two groups of lakes were defined only by the study regions (The Chagan lake group is in the Songyuan, the Yueliang lake group in the Baicheng. The Yuelaing lake group also comprise relatively high salt lake such as Talahong (TLH) as well as low salt lake (YLL). This manuscript is to assess the dynamic of individual CDOM fluorescent component under seasonal variation contained in both fresh and saline lakes in the western part of Jilin province rather than compare fresh and brackish water CDOM. The differences of fluorescent components in CDOM between fresh and saline lakes during different seasons should be studied in further study, particularly with larger saline gradients as presented in Song et al., (2013).

- The description of the fluorescent components is often not only confusing (e.g. sometimes the PARAFAC component C3 is also referred to as "protein-like peak T" (as defined by other authors) but sometimes also wrong. In general the authors might improve their understanding of PARAFAC fluorescent components, for example the components should be called "fluorescent components" rather than "fluorescence components". Furthermore, the PARAFAC components should always be referred to as "components" and not as "peaks". Also, the claim that the two protein-like components "[…] consist of two dissolved amino acids, i.e., tryptophan and tyrosine […]" is wrong, they should be rather referred to "tryptophan-like" or "tyrosine-like".

Response: Thank you for your suggestions. In order to understand of EEM-PARAFAC fluorescent components, the "fluorescence components" should be replaced by "fluorescent components". The "tryptophan" or "tyrosine" components should be replaced by "tryptophan-like" or "tyrosine-like" components. The "PARAFAC peaks" should be replaced by "PARAFAC components" throughout the paper. The reviewer's comments have been incorporated in the revised manuscript.

- **For some** data statistics need to be clarified. For example, the seasonal changes of fluorescence intensities of the four PARAFAC components is described in the text as “increasing” and “decreasing”, partly also “significantly”, though Figure 5a hardly reveals any obvious seasonal changes for most of the components.

Response: Thank you for your suggestion. Figure 5a and b presented seasonal average and seasonal variation of the four EEM-PARAFAC components in the revised manuscript. The total fluorescence intensity is defined as the sum of fluorescence intensities of different fluorescent components. For Figure 5a, the total fluorescence intensities varied at different seasons can not be revealed. We are sorry for making the error. When the data were used *t*-test again, partly there were no significant (*t*-test, $p>0.05$).

On page 5737 in line 16, the contents “The total fluorescence intensity significantly varied from 2.54 ± 0.68 nm^{-1} in June to 1.93 ± 0.70nm^{-1} in August 2013, and then increased to 2.34 ± 0.92 nm^{-1} in February and reduced to the lowest 1.57 ± 0.55nm^{-1} in April 2014.” should be replaced by “The total fluorescence intensity varied from 2.54 ± 0.68 nm^{-1} in June to 1.93 ± 0.70 nm^{-1} in August 2013, and then increased to 2.34 ± 0.92 nm^{-1} in February and reduced to the lowest 1.57 ± 0.55 nm^{-1} in April 2014.”

Responses to specific comments

Page 5727

Line1: DOM consists of several humic acids rather than only one. Further it contains “proteins”, not “proteinaceous”.

Line2: skip “array” in the sentence

Response: Thank you for pointing out the mistakes. The contents “Dissolved organic matter (DOM), a heterogeneous mixture of humic acid, proteinaceous and carbohydrates, plays an array of important roles in aquatic ecosystems.” should be replaced by “Dissolved organic matter (DOM), a heterogeneous mixture of humic acids, proteins and carbohydrates, plays an important role in aquatic ecosystems.”

Line3: CDOM absorbs light of ultraviolet and visible light (not only within the blue region, though the absorption here is in fact typically higher than at the longer wavelengths of visible light).

Response: Thank you for pointing out the mistake. The contents "……absorbs light energy in the UV (280–400 nm) and blue region of the spectrum" should be replaced by "……absorbs light energy in the UV (280–400 nm) and visible region of the spectrum".

Line9: autochthonous CDOM does not only originate from plant materials but also from a range of other organisms (e.g. algae, microorganisms)

Line10: use "terrestrially imported substances"

Responses: Thank you for pointing out the mistakes. The contents "…CDOM originates from the degradation of plant materials and terrestrial imported substance" should be replaced by "…CDOM originates from the degradation of plant materials and other organisms and terrestrially imported substance".

Line12: use "making it difficult to isolate…"

Response: Thank you for pointing out the mistakes. The contents " …..which makes it difficult to isolate…" should be replaced by "…, making it difficult to isolate…".

Line14: the terms "OACs" and "SFS" are never again used in the paper. I suggest to skip these abbreviations

Response: Thank you very much for pointing out the mistakes. These abbreviations "OACs" and "SFS" should be skipped.

Line23: skip "and then"

Response: Thank you for pointing out the mistake. The contents "…and then…" should be skipped.

Line26: use "resulting in an emission of lower energy"

Response: Thank you for pointing out the mistake. The contents "…resulting in the emission energy lower than …" should be replaced by "…resulting in an emission of lower energy…"

page 5728

Line 6: abbreviation of EEM has already been mentioned before

Response: Thank you for your suggestion. The contents "the fluorescence excitation-emission matrix s (EEMs)" should be replaced by "EEM spectroscopy".

Line 8: better write "EEM spectroscopy" than "EEMs"

Response: Thank you for your suggestion. The contents "EEMs" should be replaced by "EEM spectroscopy".

Line 11ff: though sometimes equalized in literature, "allochthonous" is not necessarily "humic-like", neither is "autochthonous" necessarily "protein-like".

Line 12: not only CDOM from coastal environments have been investigated, also from freshwater and other systems

Line 15: re-formulate the sentence, e.g. "…of marine, freshwater and ice-water ecosystems as well as snow"

Responses: Thank you for pointing out the mistakes. The contents "…distinguish allochthonous (humic-like fluorophores) and autochthonous (protein-like fluorophores) CDOM source in coastal environment… and to investigate the dynamics of various aquatic ecosystem including marine, freshwaters and snow, ice-water systems" should be replaced by "…distinguish allochthonous and autochthonous CDOM source and to investigate the dynamics of coastal environment, marine, freshwater and ice-water ecosystems as well as snow melting water".

Line17: skip repetitive content (see line 19ff, page5727)

Response: Thank you for your suggestion. The contents "…two main fluorescence components, i.e., humic-like and protein-like (Peaks B and T) substance …" should be replaced by "…the humic-like and protein-like substances…".

Line 26ff: skip parts that are repetitive to line 21ff

Response: Thank you for your suggestion. The contents "Stedmon et al. (2003) introduced parallel factor analysis (PARAFAC) to decompose EEMs into individual fluorescent components. Their investigation identified…" should be replaced by "Stedmon et al. (2003) introduced PARAFAC and identified…"

Page 5729

Line 3: use "PARAFAC modelling" and "correlations of the fluorescent components…"

Response: Thank you for suggestion. The contents "…by PARAFAC model and analyzed the correlations the fluorescence components…" should be replaced " … by PARAFAC modeling and analyzed the correlations between the fluorescent components…".

Line 8: use "low average SUVA254"

Response: Thank you for pointing out the mistake. The contents "small average SUVA254" should be replaced "low average SUVA254".

Line 9: use "indicate"

Line 9: S_R indicates "low average DOM molecular weight"

Response: Thank you for your suggestion. The contents "…indicating large portion of lower molecules …" should be replaced by "…, which indicate large portion of low average DOM molecular weight…".

Line 11: "…may be stored…"

Response: Thank you for pointing out the mistake. The contents "…may store…" should be replaced by " …may be stored…".

Line 12: skip "furthermore"

Line 12: maybe better use "Therefore" instead of "to achieve this goal"

Response: Thank you for your suggestion. The word "furthermore" should be skipped.

The contents "to achieve this goal" should be replaced by "Therefore".

Line 14: "at different seasons"

Response: Thank you for your suggestion. The contents "… in different seasons…" should be replaced by "…at different seasons…".

Line 17: "assess the dynamics of individual fluorescent components under seasonal variations"

Response: Thank you for your suggestion. The contents "…assess the dynamic of individual fluorescence component of CDOM with seasonal variations…" should be replaced by "…assess the dynamic of individual fluorescent components under seasonal variation…".

Line 19: unclear which factors are related to each other. I suggest "[…] link CDOM fluorescence intensities, absorption coefficients, DOC concentrations and salinity to each other."

Response: Thank you for your suggestion. The contents "…link CDOM fluorescence intensity and the absorption coefficients, DOC concentration and salinity as well." should be replaced by"… link CDOM fluorescence intensities, absorption coefficients, DOC concentrations and salinity to each other."

Materials and methods

Response to general:

- When were the lakes covered with ice? How were samples taken during ice coverage?

Response: The lakes were covered with ice in late November every year. During ice coverage, the samples were collected by using an ice drilling. After the ice layer was drilled a hole, the underlying water was coming up. The ice shavings was collected in the plastic bags and the lake water was collected in the plastic bottles.

- were any other parameters measured that could have affected CDOM measurements? (e.g. iron content)

Response: The existing data in this study can not explain the question. The other parameters e.g. iron content that could have affected CDOM measurements will be measured in the future study.

Line 25: use "two groups of lakes…"

Response: Thank you for pointing out the mistake. The contents "Two group of lakes…" should be replaced by "Two groups of lakes…".

Line 27: use "with a relatively high mean salinity of…"

Response: Thank you for pointing out the mistake. The contents "with the mean high salinity" should be replaced by "with a relatively high mean salinity of…".

Page 5730

Line 4: skip "related to"

Line 4: skip "for the study region"

Response: Thank you for your suggestion. The contents "…related to…" and "For the study region” should be skipped.

Line 5: “the average annual precipitation is about 1790mm”

Response: Thank you for your suggestion. The contents “…the average annual precipitation is 391mm” should be replaced by “…the average annual precipitation is about 391mm”.

Line 7: better use “agricultural catchment land use”

Response: Thank you for your suggestion. The contents “…agricultural practice…” should be replaced by “…agricultural catchment land use…”.

Line 10: “…field campaigns in June and August 2013 as well as in February and April 2014” might be easier to read

Response: Thank you for your suggestion. The contents “…four field campaigns in July, August 2013 and February, April 2014, respectively” should be replaced by “…four field campaigns in June and August 2013 as well as in February and April 2014, respectively.”.

Line13: have the samples been stored unfiltered?

Response: The water samples were filtered and then stored before analysis.

Page 5731

Line 2: skip “were”

Line 2: Whatman GF/F filters have a 0.7 µm nominal pore size

Response: Thank you for pointing out the mistakes. We are sorry for making the mistakes. The contents “…water samples were filtered through pre-combusted 0.45 µm Whatman GF/F filters and then were measured…” should be replaced by “…water samples were filtered through 0.45 µm filters and then measured…”.

Line 20: “…where the absorbance of CDOM…”

Response: Thank you for pointing out the mistakes. The contents “…that the absorbance of CDOM…” should be replaced by “…where the absorbance of CDOM…”.

Line 24: equation: Why has 440nm been used as reference wavelength?

Response: The wavelength 440 nm has been chosen as reference wavelength in other literatures (Babin et al., 2003; Bricaud et al., 1995; Song et al., 2013; Zhang et al., 2010). That is because CDOM absorption coefficients become exponentially increase in the shorter wavelength regions (280-400nm) than 440nm.

Page 5732

Line 4: The slope ratio S_R by Helms et al., 2008 further indicates the molecular weight and photo-bleaching of DOM

Response: Thank you for pointing out the mistakes. We are sorry for making the mistakes. The contents "…indicate the contribution of different sources in terms of their molecular weights" should be replaced by "…indicate the molecular weight and photo-bleaching of CDOM".

Line15: I would not refer the inner-filter effect as "the major problem with EEMs measurement of CDOM". It highly depends on DOC concentration and the contribution of humic and fulvic acids.

Response: Thank you for your suggestions. The major problem with EEMs measurement of CDOM depends on DOC concentration and the contribution of humic and fulvic acids. The contents "The inner-filter effect is the major problem with EEMs measurement of CDOM because of …" should be replaced by "The inner-filter effect, which results from reabsorption and excitation of the fluorescence itself…"

Line 17: "[…] can reduce the fluorescence intensity by 5%"

Response: Thank you very much for your suggestion. The contents "…can reduce the intensity of fluorescent component by 5%" should be replaced by "…can reduce the fluorescence intensity by 5%".

Line 23: If written in this form, the formula needs more brackets surrounding (Aex + Aem)!

Response: Thank you for your suggestion. The formula "(Aex+Aem/2)" should be replaced by "(Aex+Aem)/2 ".

Line 25: rather use "respective" than "current"?!

Response: Thank you for your suggestion. The word "current" should be replaced by "respective".

Line 27: I guess the Milli-Q water Raman peak was "measured" daily rather than “collected”?

Response: Thank you for your suggestion. The contents “…collected daily” should be replaced by “…measured daily”.

Page 5733

Line 4: PARAFAC modeling decomposes the CDOM fluorescence signal into separate fluorescent signals

Response: Thank you for pointing out the mistake. The contents “…decompose EEMs of the complicate mixture CDOM into individual fluorescence groups” should be replaced by “…decompose the CDOM fluorescence signal into separate fluorescent signals”.

Line 18: matrices

Line 18: Excel (Microsoft office)?

Response: Thank you for pointing out the mistake. The word “matrixes” should be replaced by “matrices”. The “Excel file” refers to “Microsoft office Excel file”.

Line 19: step (2) and (3) described here are repetitive to line 3ff of page 5734 and should be deleted here!

Response: Thank you for your suggestion. The paragraph “In our study…carried out in the “N-way DOMFluor toolbox for MATLAB” should be skipped.

Page 5734

Line 2: Andersson et al.

Response: Thank you for pointing out the mistake. The contents “…Andersso et al., 2000” should be replaced by “…Andersson et al., 2000”.

Line 6: “the Rayleigh bands do not represent DOM fluorescence”

Response: Thank you for your suggestions. The contents “…do not describe DOM fluorophore in the area” should be replaced by “…do not represent DOM fluorescence in the area”.

Page 5735

Line 5: “[…] the set of samples was pooled together […]”

Response: Thank you for your suggestions. The contents “When the whole set of samples were pooled together…” should be replaced by “When the set of samples was pooled together…”.

Line 7ff: better write e.g. “…whereas the other three seasons exhibit relatively constant values of approximately 0.40 PSU.”

Response: Thank you for your suggestions. The contents “The salinity for lakes with other three seasons was almost the same, which was about 0.40 PSU” should be replaced by “…whereas relatively constant values (around 0.40 PSU) were exhibited in other three seasons”.

Line 11: Why does turbidity decrease with increasing rainfall?

Response: When the particulate substances in the lakes are resuspended with the wind-induced mixxing, the turbidity of waters can correspond to change. To the shallow lakes in the Songnen Plain, the wind is relatively weak from July to August every year. Though terrestrial matters are imported to water bodies through rainwash with rainfall increasing, the particulate substances deposited immediately. Also, the turbidity of pure rainwater is about 0. Therefore, the turbidity reduced to 63.13 ± 31.21 NTU in August 2013 with rainfall increasing.

Line 10ff: For easier reading I suggest to skip the values here and refer to the corresponding table.

Response: Thank you for your suggestions. The contents “The highest turbidity was present in June 2013 with 166.20 ± 108.73 NTU, then reduced to 63.13 ± 31.21 NTU in August 2013 with rainfall increasing, and then reduced to the lowest 21.33 ± 15.87 NTU in February 2014. Compared with the turbidity in February 2014, the turbidity had almost no change in April 2014 with 22.24 ± 16.42NTU.” should be replaced by “The highest turbidity was present in June 2013, then reduced in August 2013, and the lowest value was recorded in February 2014. Compared with the turbidity in February 2014, the turbidity had almost no change in April 2014 (Table 1).”

Which of these changes are statistically significant? What about the average salinities of the two groups of lakes? Are there any significant seasonal changes on pH? This might be important since pH can affect fluorescence (e.g. see Reynolds 2003: “Rapid and direct determination of tryptophan in water using synchronous fluorescence spectroscopy")

Response: Thank you for recommending the reference "Reynolds 2003: "Rapid and direct determination of tryptophan in water using synchronous fluorescence spectroscopy". Table 1 showed that the values about water quality parameters pH, salinity, turbidity at different seasons for both fresh and saline lakes. Since there are no significant seasonal changes on pH which can affect fluorescence. The manuscript is to assess the dynamics of individual fluorescent components under seasonal variation for both fresh and saline lakes in the western part of Jilin province in Northeast China rather than compare the difference in fluorescence between fresh water and saline water.

Line 15ff: I would rather say "fluorescent components" instead of "fluorescence peaks". Further, to avoid confusion, I would rather compare the fluorescent components of this study with those of other studies only once in the beginning but then stick with the terminology given in this study (e.g. use "C1", "C2" etc. throughout the manuscript instead of switching to "A" or "C").

Response: Thank you for your suggestions. The contents "fluorescence peaks" should be replaced by "fluorescent components" throughout the manuscript. We would use "C1, C2, C3 and C4" throughout the manuscript rather than "A", or "C" or "B" or "T". The reviewer's comments have been incorporated in the revised manuscript.

However, in here, based on the EEMs "peak picking" technique, the fluorescence peaks e.g. humic-like A and C and protein-like B and T was firstly defined by Coble et al. (1996), which is only cited here in the manuscript.

Line 18: see General Comments

Response: Thank you for your suggestion. The contents "The humic-like component is a complex mixture of aromatic and aliphatic compounds-fulvic acids and humic acids, …i.e., tryptophan and tyrosine…." should be replaced by "The humic-like component is a complex mixture of aromatic and aliphatic compounds- humic-like acids from terrestrial substances, and aquatic humic-like substances of phytoplankton origin…i.e., tryptophan-like and tyrosine-like substances…".

Line 23: I suggest reconstructing the structure of the sentence, e.g. "As an example,

Figure 2 displays examples of EEMs of lake Xindianpao at the four different seasons."

Response: Thank you for your suggestions. The contents "Taking Xindianpao as an example with different seasons (Fig. 2)" should be replaced by "As an example, Figure 2 displays examples of EEMs of lake Xindianpao at the four different seasons".

Page 5736

Line 3ff: this paragraph is partly redundant to the second last paragraph of page 5734 and should only be described in the method-section.

Response: Thank you for your suggestions. The contents "To determine the appropriate number of PARAFAC components, the split-half validation procedure was executed to verify whether the model was valid by comparing the emission and excitation loadings from each half (Stedmon and Bro, 2008)" should be placed in the Method section on Page 5734 in Line 11.

Line 18ff: I did not find evidence in literature that dedicates component C1 to phytoplankton degradation, rather relations to marine humic-like substances, waste water treatment or agricultural catchment land use

Response: In marine environment, C1 displayed similar fluorescence peaks to marine humic-like substances (Coble et al. 1996). For inland waters, the component C1 is related to phytoplankton degradation which is found by Zhang et al. (2009) (The contribution of phytoplankton degradation to chromophoric dissolved organic matter (CDOM) in eutrophic shallow lakes: Field and experimental evidence. Water Res. 43: 4685–4697).

Line 22ff: I would not compare C2 to C1 like in "Compared with C1 […] component shifted to the red spectral region".

Response: Thank you for your suggestions. In order to differentiate component C1 from C2 which are both related to the humic-like components, we use "Compared with C1 […] component 2 shifted to the red spectral region". The contents "Compared with C1 […] component 2 shifted to the red spectral region" should be replaced by "Component 2 had a maximum excitation (at 255 and 350 nm) and emission (at 460 nm) characteristics…”.

Line 25: maybe use “C3 resembles the tryptophan-like component as found by (author name).

Response: Thank you for your suggestions. The contents “Component 3 demonstrated two excitation maxima (at 225 and 290 nm) and one emission maximum (at 360 nm), which is considered to be similar to tryptophan.” should be replaced by “Component 3 demonstrated two excitation maxima (at 225 and 290 nm) and one emission maximum (at 360 nm), which resembles the tryptophan-like component as found by Baker et al. 2004 (Baker et al., 2004; Hudson et al., 2007).”

Page 5737

Line 4: maybe better write “In this study not all of the four components were present in all of the samples.”

Response: Thank you for your suggestions. The contents “However, these results from our study do not mean all the four components were shown in every water sample.” should be replaced by “In this study, not all of the four components were present in all of the samples.”

Line 13ff: The message here is unclear. Maybe better write e.g. “At all four seasons the fluorescent component C2 contributed less to total fluorescence than C1”?

Response: Thank you for your suggestions. This means the average fluorescence intensity of C2 is the lowest than the other three components (C1, C3 and C4). The contents “The lowest C2 intensity represented only a small portion of CDOM…” should be replaced by “At all four seasons, the fluorescent component C2 contributed less to total fluorescence than the three others…”

Line 19ff: Message unclear, please reconstruct the sentence

Response: Thank you for your suggestions. The contents “ In June 2013, the intensities of four fluorescence components (0.75 ± 0.17 (C1), 0.32 ± 0.06 (C2), 0.69 ± 0.24 (C3), 0.77 ± 0.20 nm (C4)) exhibited the similar trends to the whole set of samples, but higher than the average except the C2 0.32 ± 0.06nm^{-1}” should be replaced by “ In June 2013, the intensities of four fluorescent components (0.75 ± 0.17 (C1), 0.32 ± 0.06 (C2), 0.69 ± 0.24 (C3), 0.77 ± 0.20 (C4) nm^{-1}) exhibited similar trends to that for the pooled data set. These values were higher than the seasonal average except C2 (0.32± 0.06 nm^{-1}). These values were higher than the seasonal average except C2 (0.32 ± 0.06 nm^{-1})."

Line 23: what is meant with "others"?

Response: The "others" means "microbial activities". The contents "This can be explained by enhanced activities from plant degradation and others." should be replaced by "This can be explained by enhanced activities from plant degradation and microbial activities."

Line 27: Is there a significant difference? According to Figure 5a there is none.

Response: Compared with the fluorescence intensity in June, the protein-like components (C3 and C4) showed an obvious difference in August 2013. These two values from C3 and C4 are never remained the same, which can be exhibited in Figure 5.

Page 5738

Line 2: this refers to Figure 6b. Further, to me it is unclear if Figure 6 is from Cheng et al., 2010 or from this study.

Response: Thank you for putting forward the questions. The results in Figure 6b are from our study, which are consistent with Cheng et al. (2010). The contents "The fluorescence intensity of rainwater was mostly characterized by protein-like components and lower humic-like concentration (Fig. 7b) (Cheng et al., 2010)." should be replaced by " In accordance with Cheng et al. 2010, the fluorescence intensity of rainwater for this study was largely characterized by protein-like components. Also the rainwater CDOM contains much lower humic-like concentration (Fig. 7b) (Cheng et al., 2010)."

1st paragraph: If rainwater contains a lot of protein-like substances why does the protein signature of the lake EEMs decrease during rainfall? Is the fluorescence weak and weighted out by the dilution effect?

Response: Thank you for putting forward the questions. Though the rainwater was dominated by protein-like components, the fluorescence intensity was very weak. The CDOM fluorescent components for lake water were importantly affected by both the degradation of plant materials and terrestrially imported substance in August 2013. The fluorescence intensities of the four fluorescent components were much higher than that of the rainwater CDOM. These fluorescence intensities of components in ~~CODM~~ from lake water may became weaker with the rainfall increasing compare to that in June 2013. The intensities of the other three components but C2 decreased because of dilution with a lot of rain.

Line 12: maybe you could add a quotation for the statement “strong biological activities would be prohibited”. Ice cover solemnly does not prevent biological activities.

Response: The ice cover reduces light penetration and restricts gas exchange between the underlying water and atmosphere. Therefore, the biological activities in lake waters became weak at the low tempreture and low light level when ice was covered (Thomas K., 1983. Under Landfast ice. Arctic, 36, 328-340; Wharton, R. A., Jr., McKay, C. P., Clow, G. D., and Andersen, D. T., 1993. Perennial ice covers and their influence on Antarctic lake ecosystems. Antarct. Res. Ser. 59, 53–70; Uusikiv, J., Vahatal, A.V., Granskog, M.A., Sommaruga, R., 2010. Contribution of mycosporine-like amino acids and colored dissolved and particulate matter to sea ice optical properties and ultraviolet attenuation. Limnology and Oceanography, 55(2), 703–713.).

Line 12f: why would C1 and C3 accumulate simultaneously? If “strong biological processes” are really prohibited how can C3 accumulate? What about C4? Where can an increase in fluorescence come from in winter during ice coverage? (autochthonous production? groundwater inflow?) Are some components preferentially “taken up” into the ice during ice formation?

Response: Thank you for putting forward the questions. In cold winter, the surface waters formed a thick layer of ice covering the lake waters after frozen. Because the ice cover reduces light penetration and restricts gas exchange between the underlying water and atmosphere, the strongly biological activities in lake waters would be prohibited at the low temperature and low light level when the ice was covered. During ice formation, C4 was preferentially taken up into the ice. Also, the dissolved matters were left in the underlying surface waters (Stedmon et al., 2007). Therefore, the C1 and C3 in the waters beneath the ice layers would cumulate simultaneously.

Line 16: here it is unclear what is meant by "ice DOM fluorescence". Were samples from ice taken as well? If so, what are the properties of the ice DOM compared to lake DOM?

Response: Thank you for putting forward the question. The samples from ice were taken as well when the water samples were collected in Feburary 2014. Compared with lake water DOM, The "ice DOM fluorescence" was dominated by tyrosine-like component and the fluorescence intensity was very weak.

Line 18ff: In how far is it not surprising that C4 fluorescence intensity beneath the ice was reduced?

Response: When the ice was drilled through, the lake water was coming up and then collected in the plastic bottles. The intensity of C4 for the collected lake water beneath ice layer was reduced.

Line 21ff: message unclear. what is meant with "similar trends with the whole"?

Response: Thank you for putting forward the question. In April 2014, the intensities of four fluorescent components showed similar seasonal trends though these values were much lower than the average.

Line 23ff: Unclear interpretation. What exactly is tried to be explained? Is the ice DOM expected to exhibit the same DOM composition as the lake DOM and therefore just dilutes lake DOM without changing fluorescent component proportions when the ice melts in spring?

Response: Thank you for your suggestion. In April 2014, the intensities of four fluorescent components was lower than the seasonal average when the set samples as a whole. This was because when the ice in the lakes melt into water with the weather warming, and the biological degradation and human activity was weak. Therefore, CDOM in lake waters was diluted and the fluorescence intensity of the four fluorescent components reached to the lowest, respectively. The ice DOM was characterized by tyrosine-like component (C4) (Figure 6a), while the lake DOM was dominated by both humic-like (C1 and C2) and protein-like (C3 and C4) components.

It can be demonstrated that when the ice melts in spring, the fluorescent component proportions changes from Feburary to April 2014 (Figure 5a).

Line 26ff: skip last paragraph since it will be mentioned in the conclusions again anyway.

Response: Thank you for your suggestion. The contents "Compared with previous works…and ice-melt water." should be placed in Line 23 on page 5741.

Page 5739

Line 4: there are no salinity values in Table 3.

Response: Thank you for pointing out the mistakes. We are sorry for making the mistakes. The content " …salinity…" should be skipped.

Line6ff: DOC-concentration: I suggest giving a short explanation for seasonal DOC dynamics.

Response: Thank you for your suggestions. The highest DOC concentration 55.04 ± 20.00 mg L^{-1} was present in Feburary 2014, wheras the other three seasons exhibit relatively constant values of approximately 30 mg L^{-1}, demonstrating a seasonal dynamic that can be attributed to hydrological, climatic and landscape variation (Song et al., 2013). This is because the DOC was accumulated when lakes freeze in winter, which left DOC in the lake waters, resulting in a high DOC concentration in the underlying water (unpublished material).

Line 9: … was used as a proxy…

Response: Thank you for pointing out the mistakes. We are sorry for making the mistakes. The contents "…was used as an proxy…" should be replaced by "…was used as a proxy…".

Line12ff: partly repetitive

Response: Thank you for your suggestions. The contents "The highest CDOM absorption coefficients a(350) 6.36 ± 2.17m^{-1}, a(280) 34.62 ± 11.54m^{-1}, a(254) 52.88± 18.13m^{-1} were present in February 2014, corresponding to the highest DOC concentration of 55.04 ± 20.00mg L^{-1}." should be replaced by "The highest CDOM absorption coefficients a(350), a(280), a(254) were present in February 2014, corresponding to the highest DOC concentration."

Line 15: explanation for DOC concentration increase in winter questionable. Might there be an increase in DOC concentration because of the water freezing (leaving DOM in the liquid phase) rather than DOC-expel from the ice?

Response: Thank you for your suggestions. The contents "This can be attributed to the accumulated dissolved organic carbon when lakes freeze in winter, which expels DOC from ice…" should be replaced by "This can be attributed to the accumulated dissolved organic carbon when lakes freeze in winter, which leaves DOC in the liquid phase…".

Line 18: "S_R" might better be called slope ratio (Helms et al. 2008)?

Response: Thank you for your suggestions. The content "…the spectral ratio (S_R)…" should be replaced by "…the slope ratio (S_R)…".

Line 21: low values of S_R indicate high molecular weight! Therefore the interpretation here is wrong.

Response: Thank you for pointing out the mistakes. We are sorry for making the mistakes. The contents "…which indicated the higher activities of biology and others resulting in decomposition of higher molecule carbon into lower molecule." should be replaced by "…which indicated the higher activities of biology and terrestrially imported substance through rainwash resulting in higher average molecular weight in DOC".

Line 21: what is meant with "high activity of biology and others"?

Response: The "high activity of biology and others" means the CDOM in lakes was originated from phytoplankton degradation and terrestrially imported substance and microorganisms in August 2013.

Page 5740

Line 1: I would not claim that C1 and C2 are "controlled by common sources". They both might derive from terrestrial sources and maybe e.g. underlie common processing mechanisms leading to similar dynamics?

Response: Thank you for your suggestions. The component C1 originated from phytoplankton degradation and C2 derived from terrestrially imported substance. The "two humic-like components were controlled by common sources" maybe indicate a common processing mechanisms leading to similar dynamics (Zhang et al., 2010, 2011).

Line 3: what is meant with “other human waste”?

Response: The “other human waste” means organic pollutants derived from domestic, agricultural and industrial sewerage contribute to increase the DOC concentration in lake waters which were characterized by the tryptophan-like (T) component (Baker et al., 2004).

Line 5: again, a non-existing correlation between the fluorescent components among the four seasons does not necessarily indicate different sources.

Response: Thank you for pointing out the mistakes. The contents “However, almost no correlation between humic-like peaks and protein-like peaks shows that the two components were derived from different sources.” should be replaced by “However, there is almost no correlation between humic-like components and protein-like components.”

Line 7: What is meant with steady and labile fluorescence?

Response: The “steady and labile fluorescence” refers to the changes in the ratio of humic-like : protein-like substances and fluorescent : nonfluorescent DOC (Henderson et al., 2009).

Line 10ff: Conclusion unclear. High DOC concentrations do not necessarily indicate pollution. Therefore a correlation between DOC concentration and C3 does not designate C3 is an indicator for pollution. Anyway, in my opinion C3 of this study has a quite high emission fluorescence peak and is marginally comparable to tryptophan (which lies according to literature between 340-350 Em wavelength (Baker 2004, Coble 1996).

Response: Thank you for your suggestions. The contents “A weak relationship ($R^2 = 0.42$) (Fig. 7d) was found between DOC and component 3 from the decay of plants through microbial activity or the pollution from human and animal wastes, indicating that the component 3 of the CDOM fluorescence (protein-like peak T) can be used to detect water pollution (Baker et al., 2004).” should be replaced by “The component C3 of the CDOM fluorescence (tryptophan-like peak) can be used to detect water

pollution (Baker et al., 2004; Hudson et al., 2007). A weak relationship ($R^2 = 0.42$) (Fig. 7d) was found between DOC and component 3 from the decay of plants through microbial activity or the pollution from human and animal wastes."
C3 (Ex/Em=225(290)/360 nm) has a longer emission wavelength compare to the study from Baker et al. 2004 and Coble 1996, but is almost consistence with the study from Hudson et al. (2007).

Are there any other measurements performed on the sampling sites to measure pollution (e.g. phosphate concentrations)?

Response: Thank you for your suggestion. The other measurements e.g. phosphate concentrations to indicate pollution should be investigated in future study. It has been demonstrated that the tryptophan-like fluorescence intensity strongly correlated with phosphate concentration (Baker et al., 2004). Therefore, the tryptophan-like fluorescence intensity can be applied to monitor water quality timely instead of the traditional approaches.

Line 24ff: parallel factor analysis itself leads to fluorescent components, it is not used to "characterize the seasonal variation of fluorescent components"!

Response: Thank you for your suggestions. The contents "In this study, the application of excitation-emission matrices fluorescence and parallel factor analysis (EEM-PARAFAC) to characterize the seasonal variation of four fluorescence components in CDOM was presented for 67 water samples collected from July 2013 to April 2014 in the semi-arid region of Northeast China." should be replaced by "In this study, the application of excitation-emission matrices fluorescence and parallel factor analysis (EEM-PARAFAC) to characterize four fluorescent components under seasonal variation in CDOM was presented for 67 water samples collected from June 2013 to April 2014 in the semi-arid region of Northeast China."

Page 5741:

Conclusion general: should be adapted to the revision of the paper. Furthermore, the importance of this study for a better understanding of freshwater/brackish water CDOM dynamics should be outlined.

Response: Thank you for your suggestions. The conclusions should be adapted to the revision of the paper. The contents "In this study, the application of excitation-emission matrices fluorescence and parallel factor analysis (EEM-PARAFAC) to characterize the seasonal variation of four fluorescence components…..Also, the study was to assess the dynamics of individual fluorescent components of CDOM with seasonal variations in both fresh and brackish waters rather than compare fresh water and brackish water CDOM." should be replaced by "In this study, the application of EEM-PARAFAC to characterize four fluorescent components under seasonal variation in CDOM was presented with 67 water samples collected from June 2013 to April 2014 in the semi-arid region of the Songnen Plain. Two humic-like and two protein-like components were identified using PARAFAC model. The average fluorescence intensity of the four components differed under seasonal variation from June 2013 to April 2014. The highest C1 1.02 nm^{-1} was presented in February 2014 due to the condensed CDOM caused by ice formation in winter. Especially in summer when quantities of rainfall take place and in winter when water is frozen, the fluorescence intensity is dominated by tyrosine-like component in rain and ice-melt water. Component 1 and 2 exhibited strong linear correlation (R^2 = 0.633). There were significantly positive linear relationships between F_{max} and CDOM absorption coefficient a(254) (R^2 = 0.72, 0.46, $p < 0.01$), a(280) (R^2 = 0.77, 0.47, $p < 0.01$), a(350) (R^2 = 0.76, 0.78, $p < 0.01$) for two humic-like components (C1 and C2), respectively. A weak relationship (R^2 = 0.42) was found between DOC and component 3 from the decay of plants through microbial activity or the pollution from human and animal wastes. Most importantly, a significant relationship (R^2 = 0.931) was found between salinity and DOC. However, almost no obvious correlation was found between salinity and EEM-PARAFAC extracted components except C3 (R^2 = 0.469), though the correlation was not as strong as with DOC concentration. "

Also, these brackish and fresh water are endowed with similar geological, hydrological and climatic settings, thus we presume that similar process may control the CDOM components. The study was to assess the dynamics of individual fluorescent components of CDOM under seasonal variation for both fresh and brackish waters rather than compare fresh water and brackish water CDOM. The differences of fluorescent components in CDOM between fresh and saline lakes during different seasons should be studied in further study, particularly with larger saline gradients as presented in Song et al., (2013).

Tables and Figures

Table 1: are there significant changes in salt concentration between the seasons? If so, might there be any possible effects on CDOM absorbance/fluorescence measurements?

Response: There are significant changes in salt concentration between the seasons. When the lakes were frozen, the highest salinity 0.70 PSU was present in February 2014. Whereas, the other three seasons exhibit relatively constant values of approximately 0.40 PSU. The salinity may has some effects on CDOM absorbance/fluorescence measurements which will be studied further later.

Table 2: “max” and “min” of the “Exmax” and “Emmax” labels should be written in subscript. Instead of “Label 1” and “Label 2” I suggest writing e.g. “Components (Coble)” and add details in the table description. Further, I would rather use “secondary maxima” instead of “secondary band”.

Response: Thank you for your suggestion. The labels “Exmax” and “Emmax” should be replaced by “$\mathrm{Ex}_{\mathrm{max}}$” and “$\mathrm{Em}_{\mathrm{max}}$”.

The “Label 1 and Label 2” should be replaced by “Components (Coble) and (Zhang) and Components (Stedmon and Markager).”

The “Secondary excitation band” should be replaced by “Secondary excitation maxima”.

Table 3: DOC concentration should be in “mg L-1”. Instead of “item” I would write e.g. “sampling season”.

Response: Thank you for pointing out the mistakes. The contents “DOC mg^{-1} L^{-1}” should be replaced by “DOC mg L^{-1}”. The word “item” should be replaced by “sampling season”.

Table 4: state clearly that the values in the table represent R and the asterisks represent the p-value

Response: Thank you for your suggestion. The description "Correlation coefficients (*R*) and significance levels (*p*) of the linear relationships between CDOM absorption, DOC, salinity and fluorescence components." should be replaced by "Correlation coefficients (*R*) and significance levels (*p*) of the linear relationships between CDOM absorption, DOC, salinity and fluorescent components (the values represent *R* and the asterisks reprensent *p* value)."

Figure 2: It should be mentioned that fluorescence is in Raman units [m-1].

Response: Thank you for your suggestion. It has been mentioned that the fluorescence in Raman units [nm^{-1}] (Larsson et al., 2007; Stedmon et al., 2003).

Figure 3: What is "(1-2 left; 3-4 right") related to? Do "a)" and "b)" each display the results of one split half analysis?

Response: Thank you for pointing out the mistakes. We are sorry for making the mistakes. The content "Results from split-half analysis (1–2 left; 3–4 right) in PARAFAC models" should be replaced by "Results from split-half analysis (1–2 up; 3–4 down) in PARAFAC models". "a)" and "b)" each display the results of one split half analysis.

Figure 4: The figure description here is not correct. This figure shows the contour plots of the four PARAFAC fluorescent components (a-d) and excitation (black) and emission (red) loadings (e-h) of each component. Fluorescence is in Raman units [m-1].

Response: Thank you for pointing out the mistakes. The contents "The PARAFAC model output showing fluorescence signatures of the four fluorescence components (**a**–**d**), the contour plots represent spectral shapes of excitation and emission. The line plots represent split-half validation results (**e**–**h**): excitation (left) and emission (right) spectra." should be replaced "The PARAFAC model output shows the contour plots of the four PARAFAC fluorescent components (a-d) and excitation (black) and emission (red) loadings (e-h) of each component. Fluorescence is in Raman units: nm^{-1} (Larsson et al., 2007; Stedmon et al., 2003)."

Figure 5a: The bars showing Fmax of the seasonal average might be visually separated from the four single seasons and may be better referred to as e.g. “seasonal average” than “all samples”. The n should be given in the description. The components might be better referred to as “EEM PARAFAC components”. Further I suggest writing “The error bars represent standard deviations”

Response: Thank you for your suggestion. The seasonal average of F_{max} has been separated from the four single seasons in the revised manuscript. As is shown in the following :

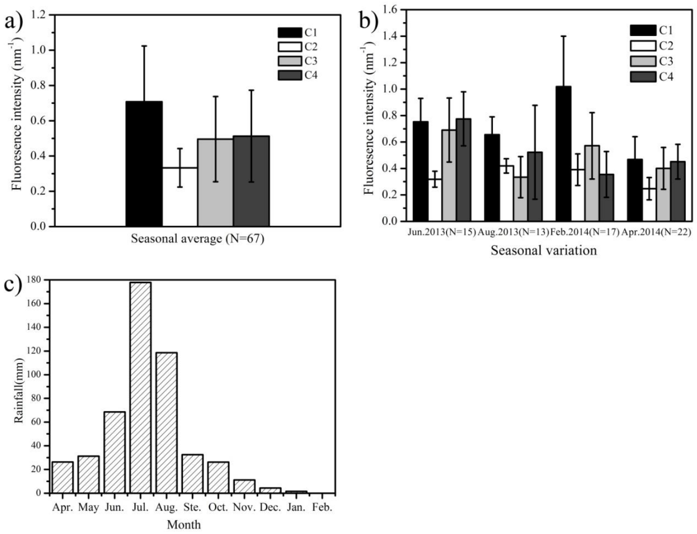

Figure 5. a) Seasonal average of F_{max} for EEM-PARAFAC components (C1, C2, C3 and C4) for lakes in the western part of Jilin province; b) Seasonal variation of the four EEM-PARAFAC components at different seasons; c) Monthly variation of rainfall for the lakes in western part of Jilin province from April 2013 to February 2014. The error bar represents standard deviations.

The contents “All samples; Jun. 2013; Aug. 2013; Feb. 2014; Apr. 2014 ” should be replaced by “Seasonal average (N=67); Jun. 2013 (N=15); Aug. 2013 (N=13); Feb.2014(N=17); Apr.2014(N=22)”.

The contents “Seasonal variation of F_{max} values of the four components…” should be replaced by “Seasonal variation of F_{max} values of EEM- PARAFAC components…”.

The content “The error bar represents SD.” should be replaced by “The error bars represent standard deviations.”

Figure 6a): what is meant with the “ice-melted water sample”?

Response: The “ice-melted water sample” showed that the fluorescence intensity of lake ice-melted water is characterized by tyrosine-like components which was very weak though. (Figure 6a).

Figure 7: What kind of test has been used?

Response: The linear regression has been been used.

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Seasonal characterization of CDOM for lakes in semi-arid regions of Northeast China using excitation-emission matrices fluorescence and parallel factor analysis (EEM-PARAFAC)

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Abstract

The seasonal characteristics of ~~fluorescence~~ components in CDOM for lakes in the semi-arid region of Northeast China were examined by excitation-emission matrices fluorescence and parallel factor analysis ~~(EEM-PARAFAC)~~. Two humic-like ~~peaks C1 (Ex/Em = 230, 300/425 nm) and C2 (Ex/Em = 255, 350/460 nm)~~ and ~~two~~ protein-like ~~B (Ex/Em = 220, 275/320 nm) and T (Ex/Em = 225, 290/360 nm) peaks~~ were identified using PARAFAC. The average fluorescence intensity of the four components differed ~~with~~ seasonal variation from June and August 2013 to February and April 2014. ~~The total fluorescence intensity significantly varied from $2.54 \pm 0.68\,\mathrm{nm}^{-1}$ in June to the mean value $1.93 \pm 0.70\,\mathrm{nm}^{-1}$ in August 2013, and then increased to $2.34 \pm 0.92\,\mathrm{nm}^{-1}$ in February and reduced to the lowest $1.57 \pm 0.55\,\mathrm{nm}^{-1}$ in April 2014. In general, the fluorescence intensity was dominated by peak C1, indicating that most part of CDOM for inland waters being investigated in this study was originated from phytoplankton degradation. The lowest C2 represents only a small portion of CDOM from terrestrial imported organic matter to water bodies through rainwash and soil leaching. The two protein-like intensities (B and T) formed in situ through microbial activity have almost the same intensity. Especially, in August 2013 and February 2014, the two protein-like peaks showed obviously difference from other seasons and the highest C1 ($1.02\,\mathrm{nm}^{-1}$) was present in February 2014.~~ Components 1 and 2 exhibited strong linear correlation ($R^2 = 0.633$). ~~There were significantly~~ positive linear relationships between CDOM absorption coefficients a(254) ($R^2 = 0.72$, 0.46, $p < 0.01$), a(280) ($R^2 = 0.77$, 0.47, $p < 0.01$), a(350) ($R^2 = 0.76$, 0.78, $p < 0.01$) and $F_{\max}$ for two humic-like components (C1 and C2), respectively. A ~~close~~ relationship ($R^2 = 0.931$) was found between salinity and DOC. However, almost no obvious correlation was found between salinity and EEM-PARAFAC extracted components except for C3 ($R^2 = 0.469$). Results from this investigation demonstrate that the EEM-PARAFAC technique can be used to evaluate the seasonal dynamics of CDOM ~~fluorescence~~ components for inland waters in semi-arid regions of Northeast China~~.~~

1 Introduction

Dissolved organic matter (DOM), a heterogeneous mixture of humic ~~acid~~, ~~proteinaceous~~ and carbohydrates, plays ~~an array of~~ important ~~roles~~ in aquatic ecosystems (Zhang et al., 2010). Chromophoric dissolved organic matter (CDOM), the colored fraction of DOM, absorbs light energy in the ~~UV~~ (280–400 nm) and ~~blue~~ region of the spectrum and inhibits the propagation of ~~ultraviolet~~ radiation. CDOM in waters also affects the transport and bio-availability of materials such as trace metals and other pollutants (Song et al., 2013), so it can be used as a proxy of water quality. In natural water bodies, CDOM originates from the degradation of plant materials and ~~terrestrial~~ imported ~~substance~~, which varies in time and space and is controlled by the structure and composition (Stedmon et al., 2003). CDOM is compositionally complex, ~~which makes~~ it difficult to isolate hydrophobic ~~and~~ hydrophilic ~~acid~~ using XAD ion-exchange resins (Aiken et al., 1992; Spencer et al., 2010). Nonetheless, some optically active components ~~(OACs)~~ of CDOM can ~~be excited by certain wavelengths and re-emit fluorescence~~ (Zhang et al., 2010) so that the fluorescence spectroscopic techniques ~~such as fluorescence emission spectrometry, synchronous fluorescence scanning (SFS) and excitation-emission matrix fluorescence spectroscopy (EEMs)~~ can be used to provide detailed information about the source and concentration of CDOM. ~~Generally, surface waters contain two key fluorescence components originated from CDOM: humic-like and protein-like substances (Coble, 1996; Fellman et al., 2010; Guo et al., 2010; Hudson et al., 2007; Stedmon et al., 2003; Zhang et al., 2010).~~

~~When a molecule absorbs light energy, a loosely held electron is excited and promoted to a higher energy unoccupied orbit, and then the CDOM fluorescence occurs when the electron returns to the ground state. Some energy is lost from the excited state to ground state by collision (fluorescence quenching) and non-radiative decay including vibrational relaxation and internal conversion, resulting in the emission energy lower than the excitation energy (the Stokes' Shift). The fluorescence signatures are dependent on the structure of chemical molecules that can absorb and re-emit light~~

~~energy as well as other factors such as temperature or pH of the solution (Hudson et al., 2007; Stedmon et al., 2003).~~

~~Traditional fluorescence techniques including fluorescence emission spectrometry and synchronous fluorescence scanning applied to study CDOM components have the drawback that the output was restricted to a linear scan (Hudson et al., 2007).~~ Recently, the fluorescence excitation-emission matrix spectroscopy (EEMs) was applied to identify CDOM components ~~which provide abundant information of~~ synchronous scan spectra in the form of contours (Coble, 1996). The ~~EEMs~~ is considered the simplest and most effective technique for studying the composition of fluorophores given its high selectivity and sensitivity to CDOM in water columns (Zhang et al., 2010). In recent years, ~~fluorescence~~ spectroscopy has been widely used to distinguish allochthonous ~~(humic-like fluorophores)~~ and autochthonous ~~(protein-like fluorophores)~~ CDOM source in ~~coastal~~ environment (Coble et al., 1998; Mayer et al., 1999; Yamashita et al., 2008, 2010; Zhang et al., 2013) and to investigate the dynamics of ~~various aquatic ecosystem including~~ marine, freshwaters and ~~snow,~~ ice-water systems (Barker et al., 2006, 2009, 2010, 2013; Coble, 2007; Hudson et al., 2007; Stedmon et al., 2007). Based on the peak positions in ~~EEM~~, two main fluorescence components, i.e., humic-like and protein-like ~~(Peaks B and T) substance~~ have been identified and investigated (DelCastillo et al., 1999; Jaffé et al., 2004). However, ~~because of overlapped fluorophores of CDOM EEMs, the~~ traditional "peak-picking" method ~~may be~~ unreliable to evaluate CDOM dynamics in aquatic ecosystems (Coble, 1996; Stedmon et al., 2003). Recently, the combined EEMs-PARAFAC (parallel factor analysis) technique has been shown to effectively decompose EEM of CDOM into independent fluorescent component and assess the source of CDOM and relationships with other water quality parameters (Broisover et al., 2009; Guo et al., 2010; Zhang et al., 2010, 2011, 2013). Stedmon et al. (2003) introduced ~~parallel factor analysis (PARAFAC) to decompose EEMs into individual fluorescent components. Their investigation~~ identified five distinct DOM components for a Danish estuary and its catchment (Stedmon et al., 2003). In coastal environments, Yamashita (2008) reported on seven components using the combined

EEMs-PARAFAC technique and assess the dynamic of individual fluorophores and relationship with salinity in Ise Bay. Zhang et al. (2011) also found three different components by PARAFAC ~~model~~ and analyzed the correlations the fluorescence components and ~~the~~ absorption coefficients of CDOM for Lake Tianmu and its catchment.

~~Compared with fresh waters, Song et al. (2013) found that the brackish waters (salinity $\geq$ 0.55 PSU) exhibited high average DOC concentration contributing significantly to the carbon budget for inland waters, small average SUVA$_{254}$ and high average S_R indicating large portion of lower molecules in DOC in the Songnen Plain. As suggested by Duarte et al. (2008) and Tranvik et al. (2009), a huge amount of DOC may store in these brackish waters~~. Therefore, it motivated us to investigate the components ~~of~~ fresh and brackish waters ~~furthermore~~ in the semi-arid region. ~~To achieve this goal~~, ~~67 water samples were collected from seven lakes in the western part of Jilin province in different seasons and~~ the absorption and fluorescence of CDOM were determined for ~~these~~ water samples. The specific objectives of this study are to: (1) characterize CDOM components contained in both saline and fresh water lakes using EEMs and their origins through the EEM-PARAFAC method, (2) assess the dynamic of individual ~~fluorescence~~ component of CDOM ~~with~~ seasonal variations, and most importantly (3) link CDOM fluorescence intensity ~~and the~~ absorption coefficients, DOC concentration and salinity ~~as well~~.

2 Materials and methods

2.1 Lakes and water sampling

The water bodies ~~to be~~ investigated in this study were located in the western part of Jilin Province, which belongs to the semi-arid part of the Songnen Plain (Song et al., 2013). Two ~~group~~ of lakes were investigated, i.e., the Chagan lake group and the Yuelianghu lake group. The Chagan lake group is made up of Lake Chagan (CGL), Xinmiaopao

(XMP), Xindianpao (XDP) and Kulipao (KLP) ~~with the mean high salinity 0.55 PSU~~. The Yuelianghu lake group mainly includes Lake Yueliang (YLL), Talahong (TLH) and Xinhuangpao (XHP) ~~with the average salinity 0.33 PSU~~ (Fig. 1). The primary economic value for these lakes is ~~related to~~ fishery, agricultural irrigation and recreation. ~~For the study region, the~~ average annual precipitation is 391 mm, but the average evaporation ~~reaches~~ to 1790 mm, resulting in water scarcity. Due to the area dominated by saline-alkali soil, the rainfall flush and agricultural ~~practice~~ can result in an increase of lake salinities. In order to characterize the CDOM fluorescence components ~~with~~ seasonal variation ~~through~~ EEMs-PARAFAC, 67 water samples were collected from the surface of the seven lakes in 1 L acid-cleaned plastic bottles for four field campaigns in J~~uly~~, August 2013 ~~and~~ February, April 2014, respectively. The collected samples were held on ice and immediately transported to the laboratory in Changchun City of Jilin province within ~~3~~h. In the laboratory, these samples were kept at 4 °C until analysis within two days. Latitude and longitude of each sample location were recorded in situ using a Trimble Global Positioning System (GPS).

2.2 Analytical procedures

To characterize the basic parameters of water quality, salinity was measured through a DDS-307 electrical conductivity (EC) meter ~~with the µS cm^{-1} (micro-Siemens/centimeter) unit at room temperature (20 ± 2 °C~~) in laboratory. Salinity was expressed ~~on the basis of~~ the UNESCO practical salinity Unit (PSU 1978). The pH was measured using a PHS-3C pH meter at room temperature (20 ± 2 °C) in laboratory. Water turbidity was determined using a Shimadzu UV-2600PC UV-Vis dual beam spectrophotometer with matching 3 cm quartz cells at room temperature (20 ± 2 °C) with Milli-Q water as the reference.

~~2.3 DOC concentration measurement~~

~~To determine DOC concentrations, water samples were filtered through pre-combusted 0.45 µm Whatman GF/F filters and then were measured using a Shimadzu TOC-5000 Analyzer and a 1.2 % Pt on silica catalyst at 680 °C. Potassium hydrogen phthalate was used as standard. The reproducibility of the analytical procedure was within 2–3 % for the current study.~~

~~2.4~~ Absorption measurement

In laboratory, all the samples were filtered at low pressure, first through a pre-combusted Whatman GF/F filter (0.7 µm) and then through a pre-rinsed 25 mm Millipore membrane cellulose filter (0.22 µm) into glass bottles. Absorption spectra of the samples were measured between 200 and 800 nm at 1 nm increments using ~~a~~ Shimadzu UV-2600PC UV-Vis dual beam spectrophotometer with a 1 cm quartz cuvette and Milli-Q water as reference. The absorption coefficient a_{CDOM} was calculated from the measured optical density (OD) of the sample using Eq. (1):

$$a_{\mathrm{CDOM}}(\lambda) = 2.303\left[\mathrm{OD}_{\mathrm{S}(\lambda)} - \mathrm{OD}_{(\mathrm{null})}\right]/\gamma \qquad (1)$$

where γ is the cuvette path length (0.01 m) and the factor 2.303 converts from base 10 to base natural logarithm transformation. Some fine particles possibly remained in the filtered solution (Babin et al., 2003; Bricaud et al., 1995), therefore it is necessary to correct for scattering by fine particles and in this case, $\mathrm{OD}_{(\mathrm{null})}$ is the average optical density over 740–750 nm ~~that~~ the absorbance of CDOM can be assumed to be zero.

A CDOM absorption spectrum ($a_{\mathrm{CDOM}}(\lambda)$) can be expressed as an exponential function (Babin et al., 2003; Bricaud et al., 1995):

$$a_{\mathrm{CDOM}}(\lambda_i) = a_{\mathrm{CDOM}}(\lambda_{\mathrm{r}})\exp\left[-S\left(\lambda_i - \lambda_{\mathrm{r}}\right)\right] \qquad (2)$$

where $a_{\mathrm{CDOM}}(\lambda_i)$ is the CDOM absorption at a given wavelength λ_i, $a_{\mathrm{CDOM}}(\lambda_{\mathrm{r}})$ is the absorption estimate at the reference wavelength λ_{r} (440 nm), and S is the spectral slope of the CDOM absorption. According to Helms et al. (2008), S is calculated by fitting a linear model to the date over a wavelength range of 275–295 nm (S1) or 350–400 nm (S2). To eliminate the inter-laboratory variability, the ratio $S_R = S1/S2$ is defined to indicate ~~the contribution of different sources in terms of their molecular weights (Zhang et al., 2010)~~.

~~2.5~~ Three-dimensional fluorescence measurement

~~EEMs~~ of CDOM were ~~measured~~ using a Hitachi F-7000 fluorescence spectrometer (Hitachi High-Technologies, Tokyo, Japan) with a 700 voltage xenon lamp. The scanning ranges ~~we used~~ were 200–450 nm for excitation, and 250–500 nm for emission. Readings were collected in ratio mode at 5 nm intervals for excitation, and at 1 nm intervals for emission, using a scanning speed of 2400 nm min^{-1}. The band-passes were 5 nm for both excitation and emission. A Milli-Q water blank of the EEMs was subtracted to eliminate the water Raman scatter peaks (McKnight et al., 2001; Stemdon et al., 2003; Zhang et al., 2010, 2011).

The inner-filter effect ~~is the major problem with EEMs measurement of CDOM because of the reabsorption of the exciting by the fluorescent itself (McKnight et al., 2001), which can reduce the intensity of fluorescent component by 5 % (Larsson et al., 2007)~~. In order to eliminate the inner-filter effect, the EEMs were corrected for absorbance by multiplying each value in the EEMs with a correction factor based on the assumption that the average path length of absorption of the excitation and emission light is one-half length of the cuvette (McKnight et al., 2001; Zhang et al., 2010). The correction function is expressed as follows:

$$F_{\text{corr}} = F_{\text{obs}} \times 10^{(A_{\text{ex}}+A_{\text{em}}/2)} \tag{3}$$

where F_{corr} and F_{obs} are the corrected and uncorrected fluorescence intensities and A_{ex} and A_{em} are the absorbance values at the ~~current~~ excitation and emission wavelengths.

Finally, the fluorescence intensities ~~in all sample~~ EEMs were normalized to the area under the Milli-Q water Raman peak (λex = 350 nm, λem = 371–428 nm) ~~collected~~ daily

(Lawaetz and Stedmon, 2009). The contour figures of the EEMs were plotted using the Matlab 10.0 software package (Math Works, Natick Massachusetts, America).

~~2.6~~ The PARAFAC modeling

~~Parallel factor analysis (PARAFAC),~~ a three-way method, is applied to ~~decompose EEMs of the complicate mixture CDOM into individual fluorescence groups and the model is written as~~ (Andersen and Bro, 2003; Stedmon and Bro, 2008):

~~$X_{ijk} = \sum_{f=1}^{F} a_{if} b_{jf} c_{kf} + \varepsilon_{ijk}$~~

~~$i = 1,\ldots,I;\ j = 1,\ldots,J;\ k = 1,\ldots,K$ (4)~~

~~where F defines the number of components in the model, X_{ijk} is the intensity of fluorescence of the ith sample at the jth emission wavelength and kth excitation wavelength. In this study, a_{if} is linearly related to the concentration of fluorescence f in a sample i and b_{jf} is proportional to the fluorescence quantum efficiency of the fth analyte at an emission wavelength j; c_{kf} is the score for the fth component at the excitation wavelength j; ε_{ijk} is the residual matrix, and i, j, k represent the variability not accounted for by the relative model. A detailed description of the assumption for PARAFAC can be found in Stedmon and Bro (2008).~~

~~In our study, the PARAFAC procedure can be outlined as below: (1) the normalized excitation-emission matrixes were combined into a single Excel file consisting of 67 worksheets, (2) the excitation wavelengths from 200 to 220 nm and the emission wavelengths from 250 to 300 nm were deleted, (3) NaN was inserted in the regions $\mathrm{Ex}-20 \le \mathrm{Em} \le \mathrm{Ex}+20$ and $2\mathrm{Ex}-20 \le \mathrm{Em} \le 2\mathrm{Ex}+20$; unit: nm, (4) PARAFAC analysis was carried out in the "N-way DOMFluor toolbox for MATLAB".~~

A number of investigators have used EEMs and PARAFAC to characterize DOM in freshwater and marine aquatic environments (Cory et al., 2005; Stedmon et al., 2005; Yamashita, 2008; Zhang et al., 2010, 2011). According to Stedmon and Bro (2008), a similar PARAFAC analysis is carried out in the present study using the DOMFluor toolbox in MATLAB with the “N-way toolbox for MATLAB” (~~Andersso~~ et al., 2000). Before PARAFAC modeling, the excitation wavelengths from 200 to 220 nm and the emission wavelengths from 250 to 300 nm were deleted because of their poor quality. Also, the Rayleigh scatter should be removed prior to PARAFAC modeling because ~~the values do not describe DOM fluorophore in the area~~. In order to remove the Rayleigh scatter, the missing values (NaN-Not a number) ~~should be~~ inserted in the regions ($\mathrm{Ex}-20 \leq \mathrm{Em} \leq \mathrm{Ex}+20$ and $2\mathrm{Ex}-20 \leq \mathrm{Em} \leq 2\mathrm{Ex}+20$; unit: nm) which are significantly influenced by the first and second order scattering from the measured spectroscopic data (Hua et al., 2007; Stedmon and Bro, 2008).

Split-half analysis is the most important method included in the PARAFAC ~~model~~, in which the EEMs are randomly divided into four groups of equal size, and then analyzed for two half splits (1–2 and 3–4 half) respectively. If the correct number of components is chosen, the excitation and emission loadings from the two groups ~~of modeling~~ should show the same shape and size (Bro et al., 1997, 1999). The fluorescence intensity of every component was represented by $F_{\max}$ (Raman unit: nm^{-1}) (Stedmon et al., 2005).

~~2.7~~ Statistical analysis

Statistical ~~analyses were~~ conducted using the SPSS 16.0 software package (Statistical Program for Social Sciences). Regression and correlation ~~analyses (paired *t* test) were~~ used to ~~calculate~~ the relationship between CDOM absorption coefficient, DOC concentration, salinity and $F_{\max}$. The difference is considered to be statistically significant when p values are less or equal to 0.05.

3 Results and discussion

3.1 Water quality conditions

The water quality parameters pH, salinity, turbidity for the 67 water samples collected from June 2013 to April 2014 in the western part of Jilin province are displayed in Table 1.~~ When the whole set of samples were pooled together~~, the waters had high pH values (mean, 8.55) and high salt contents (mean, 0.48 PSU). ~~When the lakes were frozen, the~~ highest salinity 0.70 PSU was present in February 2014~~. The salinity for lakes with other three seasons was almost the same, which was about 0.40 PSU~~. Also the water bodies were highly turbid with the mean 62.18 ± 79.07 NTU. The highest turbidity was present in June 2013~~ with 166.20 ± 108.73 NTU~~, then reduced~~ to 63.13 ± 31.21 NTU~~ in August 2013~~ with rainfall increasing~~, and~~ then reduced to~~ the lowest ~~21.33 ± 15.87 NTU~~ in February 2014. Compared with ~~the turbidity in~~ February 2014, the turbidity had almost no change in April 2014~~ with 22.24 ± 16.42 NTU~~.

3.2 EEMs characterization of CDOM

Based on the EEMs “peak picking” technique, four key fluorescence peaks can be observed in 67 water samples: two humic-like~~ (Peaks A and C)~~ and two protein-like ~~(Peaks B and T)~~ substances (Coble et al., 1996; Stedmon et al., 2003). The humic-like ~~component is a complex~~ mixture of aromatic and aliphatic ~~compounds-fulvic acids and humic acids, which were originated from terrestrial materials or algal decomposition in the waters~~. ~~While~~ the two protein-like components ~~consist of two dissolved amino acids, i.e., tryptophan and tyrosine that are originated from microbial activity~~ (Baker et al., 2004; Hudson et al., 2007). ~~Taking Xindianpao as an example with different seasons (Fig. 2), the~~ peaks comprise two humic-like fluorescence ~~components: A-humic-like~~ (Ex/Em = 220–240/410–430 nm); ~~C-humic-like~~ (Ex/Em = 300–340/410–450 nm) and~~ two protein-like fluorescence components: B~~-tyrosine-like (Ex/Em = 210–230, 270–280/310–330 nm); ~~T-tryptophan-like~~ (Ex/Em = 220–230, 280–

300/350–370 nm). ~~The measured peak intensity of these fluorescence centers is dependent on the concentration of the main fluorophores dissolved in water bodies.~~

~~To determine the appropriate number of PARAFAC components, the split-half validation procedure was executed to verify whether the model was valid by comparing the emission and excitation loadings from each half (Stedmon and Bro, 2008).~~ In split-half analysis, the 67 EEMs were randomly divided into four halves and then ~~analysized~~ for two different splits (1–2 and 3–4 half split). When the number of components was chosen to be four, the excitation and emission loadings from the output results of 1–2 and 3–4 split-half analysis ~~showed~~ largely overlapped ~~pattern~~, respectively (Fig. 3). It should be noted that for the 3–4 split-half analysis, the excitation and emission loadings of component 3 and component 4 were reversed. In fact, the output results of the split-half analysis were valid as long as the excitation and emission loadings of the fluorescence component were overlapped at the same wavelength and ~~different colors of different components~~ in the two groups only represent the order of appearance of the components according to their contributions (Stedmon and Bro, 2008).

In our study, four separate fluorescent components (Fig. 4a–d) and the excitation and emission loadings (Fig. 4e–h) of the four components identified by EEM-PARAFAC are summarized in Fig. 4 and Table 2. The first fluorescent component (C1) was a biological degradation humic-like component that displays two excitation maxima (at 230 and 300 nm) with a single emission wavelength (at 425 nm) comparable to humic-like peaks in marine and in phytoplankton degradation experiments for inland waters (Coble, 1996; Zhang et al., ~~2010~~). ~~Compared with C1,~~ the maximum ~~excitation~~ (at 255 and 350 nm) and an emission (at 460 nm) ~~of component 2 shifted to the red spectral region~~ (Fig. 4a and b), which was consistent with the humic-like peaks (A and C) defined by Coble (1996). Component 3 demonstrated two excitation maxima (at 225 and 290 nm) and one emission maximum (at 360 nm), ~~which is considered to be similar to tryptophan~~. For component 4, it is likely related to ~~tyrosine~~, which was characterized by maximum excitations at 220 and 275 nm and the emission wavelength was found at 320 nm. Components 3 and 4 represent autochthonous semi-labile CDOM associated with bacteria activity and phytoplankton degradation. Particularly, there was a shoulder at the excitation wavelength 310–330 nm in component 3 and 330–340 nm in component 4, which may be due to the residual Raman peaks in some water ~~sample~~ (Fig. 4c and d). ~~However, these results from our study do not mean all the four components were shown in every water sample~~.

3.3 Temporal distribution of PARAFAC components

As shown in Fig. ~~5~~, the average fluorescence intensity of the four components ~~differed with seasonal variation~~. When all the water samples ~~in~~ different seasons were pooled together, the average value of total fluorescence intensity was 2.05 ± 0.93 nm^{-1}, corresponding to the intensities of 0.71 ± 0.32 (C1), 0.33 ± 0.11 (C2), 0.50 ± 0.24 (C3), and 0.51 ± 0.26 nm^{-1} (C4) for different components. ~~The~~ fluorescence intensity was dominated by ~~peak~~ C1, ~~indicating that~~ most part of ~~CDOM for inland waters~~ was originated from phytoplankton degradation. ~~The lowest C2 intensity represented only a small portion of CDOM, which can be attributed to terrestrial import to water bodies. The two protein-like intensities (B and T), related to bioavailability and microbial activity of CDOM, had almost the same magnitude.~~ The total fluorescence intensity significantly varied from 2.54 ± 0.68 nm^{-1} in June to 1.93 ± 0.70 nm^{-1} in August 2013, and then increased to 2.34 ± 0.92 nm^{-1} in February and reduced to the lowest 1.57 ± 0.55 nm^{-1} in April 2014. ~~In June 2013, the intensities of four fluorescence components (0.75 ± 0.17 (C1), 0.32 ± 0.06 (C2), 0.69 ± 0.24 (C3), 0.77 ± 0.20 nm^{-1} (C4)) exhibited the similar trends to the whole set of samples, but higher than the average except the C2 0.32 ± 0.06 nm^{-1}.~~ This can be explained by enhanced activities from plant degradation and ~~others~~. Compared to the fluorescence intensity in June, the three fluorescence intensities 0.65 ± 0.14 (C1), 0.33 ± 0.16 (C3), 0.52 ± 0.36 ~~nm^{-1}~~ (C4) from samples collected in August 2013 reduced, but an increased intensity of ~~C2 (0.42 ± 0.05 nm^{-1})~~ ~~was recorded~~. Especially, ~~the two protein-like peaks showed obvious difference. From June to August 2013, the precipitation substantially increased~~ up to 180 mm in July (Fig. ~~6b~~) so that ~~the~~ flood occurred when ~~the~~ rainfall continued to increase in August. ~~The fluorescence intensity of rainwater was mostly characterized by protein-like components and lower humic-like concentration (Fig. 7b) (Cheng et al., 2010). Gradually, DOM contained in terrestrial CDOM was flushed by the rainfall to the lakes so that the C2 ($0.42 \pm 0.05\,\mathrm{nm}^{-1}$) fluorescence intensity gradually became higher but all the intensities of the other three peaks decreased because of dilution with a lot of rain and so weakened active microbial decomposition of plants.~~

The highest C1 ($1.02 \pm 0.38\,\mathrm{nm}^{-1}$) presented in February 2014 and the C2 ($0.39 \pm 0.12\,\mathrm{nm}^{-1}$) intensity ~~maintained~~ almost the same as that in August 2013. However, the two protein-like ~~peaks~~ indicated that the C3 ($0.57\pm0.25\,\mathrm{nm}^{-1}$) intensity was higher than the C4 ($0.35\pm0.17\,\mathrm{nm}^{-1}$) intensity, which was opposite to the results from other months. ~~In deep winter, the surface waters, after frozen, formed with a thick layer of ice covering the lake waters and strong biological activities in waters would be prohibited, therefore the C1 and C3 in the waters beneath the ice layers would cumulate simultaneously.~~ Obviously, the fluorescence intensity of component 1 reached to the highest value ~~of $1.02 \pm 0.38\,\mathrm{nm}^{-1}$~~. As shown in Fig. 6a, another striking feature for winter samples was that ~~the ice CDOM fluorescence~~ was dominated by the ~~protein-like C4 peaks~~, which is consistent with the finding of Barker et al. (2009, 2013) and Stedmon et al. (2007). ~~It is not surprising that the intensity of peak B for water beneath ice layer was reduced and the concentrated C3 showed higher fluorescence intensity of $0.57 \pm 0.25\,\mathrm{nm}^{-1}$~~. In April 2014, the intensities of four fluorescence components (0.47 ± 0.17 (C1), 0.25 ± 0.08 (C2), 0.40 ± 0.16 (C3), $0.45 \pm$ ~~$0.13\,\mathrm{nm}^{-1}$ (C4)~~) ~~characterized by the similar trends with the whole but lower than the average.~~ Our interpretation is that ~~the ice in the lakes melt into water with the weather warming and the biological degradation and human activity was weak, CDOM was diluted and the fluorescence intensity reached to the lowest values.~~

~~Compared with previous works by Borisover et al. (2009) and Zhang et al. (2011), our study identifies the humic-like component 1 and 2, and the two protein-like components such as tyrosine-like and tryptophan-like peaks in CDOM for lakes in semi-arid region of Northeast China with obviously seasonal characteristics. Especially in summer when~~

~~quantities of rainfall taking place and in winter when water is frozen, the fluorescence intensity is dominated by tyrosine-like peak B in rain and ice-melt water.~~

3.4 CDOM vs. EEM-PARAFAC extracted components

The ~~water quality parameters~~ DOC concentration, ~~salinity and absorption coefficient~~ are shown in Table 3. ~~For the 67 water samples, the~~ DOC concentration ranged from 10.03 to 88.15 $\mathrm{mg\,L^{-1}}$ during the study period, with an average value of $37.60 \pm 18.05\,\mathrm{mg\,L^{-1}}$, demonstrating a seasonal dynamics that can be attributed to hydrological, climatic and landscape ~~variation~~ (Song et al., 2013). Generally, the absorption coefficient a(350) was used as ~~an~~ proxy for characterizing CDOM concentration, which was $5.73\pm1.68\,\mathrm{m^{-1}}$ in June 2013, $5.82\pm0.81\,\mathrm{m^{-1}}$ in August 2013, $6.36 \pm 2.17\,\mathrm{m^{-1}}$ in February 2014, $4.17 \pm 1.49\,\mathrm{m^{-1}}$ in April 2014, respectively, with ~~an average of $5.40 \pm 1.84\,\mathrm{m^{-1}}$ when the data were pooled together~~. The highest CDOM absorption coefficients a(350) ~~$6.36\pm2.17\,\mathrm{m^{-1}}$~~, a(280) ~~$34.62\pm11.54\,\mathrm{m^{-1}}$~~, a(254) ~~$52.88 \pm 18.13\,\mathrm{m^{-1}}$~~ were present in February 2014, corresponding to the highest DOC concentration ~~of $55.04 \pm 20.00\,\mathrm{mg\,L^{-1}}$~~. ~~This can be attributed to the accumulated dissolved organic carbon when lakes freeze in winter, which expels DOC from ice, resulting in a high DOC concentration in the underlying water (unpublished material). Also, the spectral ratio (S_R)~~ of the two wavelength ranges (275–295 nm over 350–400 nm) ~~was~~ used to represent the ratio of molecular weight of humic acid and fulvic acid. The mean of spectral ratio (S_R) for all water samples was up to 1.21 ± 0.20, with the lowest 0.96 ± 0.22 in August 2013 ~~which indicated the higher activities of biology and others resulting in decomposition of higher molecule carbon into lower molecule.~~

When the whole data set ($N = 67$) was pooled together, there were significantly positive linear relationships between a(254), a(280), a(350) and $F_{\max}$ for two humic-like components (C1 and C2), respectively, but mostly such correlations were not observed for the protein-like ~~peaks~~ (Fig. 7, Table 3). These results were in accordance with ~~some published literatures~~ (Zhang et al., 2010, 2011). Components 1 and 2 were strongly linearly correlated with each other ($R^2 = 0.633$), indicating that the concentrations of the two humic-like components were controlled by common sources. There was a weak relationship ($R^2 = 0.051$) between the ~~two~~ protein-like~~ peaks (B and T)~~ because of~~ the effects of~~ rainfall in summer, ice in winter and~~ other human waste~~, which represent the complex origins of CDOM. However, almost no correlation between humic-like ~~peaks~~ and protein-like~~ peaks~~ ~~shows that the two components were derived from different sources.~~ The linkage of a fluorescence signal to DOC was very complicated because of the effects of both steady and labile fluorescence and non-fluorescence CDOM ~~with seasonal impacts such as~~ increased rainfall and algal blooms which affect the DOC concentration (Henderson et al., 2009). A weak relationship ($R^2 = 0.42$) (Fig. 7d) was found between DOC and component 3 from the decay of plants through microbial activity or the pollution from human and animal~~ wastes, indicating that the component 3 of the CDOM fluorescence (protein-like peak T) can be used to detect water pollution (Baker et al., 2004).~~

The most important finding for ~~waters with seasonal variation in the western part of Jilin province~~ is a significant relationship ($R^2 = 0.931$) ~~observed~~ between salinity and DOC (Fig. 7e), which implies that~~ the~~ prolonged sunshine duration can result in an evapo-condensed DOC concentration. Different from the findings ~~from~~ Yamashita (2008), this study did not find obvious correlation between salinity and EEM-PARAFAC extracted components ~~except~~ C3 ($R^2 = 0.469$) (Table 4 and Fig. 7d). It was because that with the sunshine duration hours increasing, the photo-degradation and microbial activities become stronger, leading to ~~the more~~ transformation of labile fluorescence CDOM into non-fluorescence CDOM component (Song et al., 2013).

4 Conclusions

In this study, the application of excitation-emission matrices fluorescence and parallel factor analysis (EEM-PARAFAC) to characterize ~~the seasonal variation of four fluorescence components in CDOM~~ was presented~~ for~~ 67 water samples collected from ~~July~~ 2013 to April 2014 in the semi-arid region of ~~Northeast China~~. Two humic-like~~ peaks (C1 Ex/Em = 230 (300)/425 nm and C2 Ex/Em = 255 (350)/460 nm) and two protein-like (B Ex/Em = 220 (275)/320 nm and T Ex/Em = 225 (290)/360 nm) peaks~~ were identified using PARAFAC model. The average fluorescence intensity of the four components differed ~~with~~ seasonal variation from ~~July~~ 2013 to April 2014. ~~In general, the fluorescence intensity was dominated by peak C1 indicating that most part of CDOM for inland waters was originated from phytoplankton degradation. The lowest C2 represents only a small portion of CDOM from terrestrial import to water bodies through rainwash and soil leaching. The two protein-like intensities (B and T) have almost the same magnitude. Especially, in August 2013 and in February 2014, the two protein-like peaks showed obviously different and the highest C1 1.02 nm^{-1} presented in February 2014.~~ Component 1 and 2 exhibited strong linear correlation ($R^2 = 0.633$)~~ indicating that the concentrations of the two humic-like components were controlled by common sources~~. There were significantly positive linear relationships between F_{max} and CDOM absorption coefficient a(254) ($R^2 = 0.72$, 0.46, $p < 0.01$), a(280) ($R^2 = 0.77$, 0.47, $p < 0.01$), a(350) ($R^2 = 0.76$, 0.78, $p < 0.01$) for two humic-like components (C1 and C2), respectively. A weak relationship ($R^2 = 0.42$) was found between DOC and component 3 from the decay of plants through microbial activity or the pollution from human and animal wastes which indicated that the components 3 (protein-like peak T) can detect lake pollution derived from fluorescence CDOM. Most importantly, a close relationship ($R^2 = 0.931$) was found between salinity and DOC. However, almost no obvious correlation was found between salinity and EEM-PARAFAC extracted components except C3 ($R^2 = 0.469$), though the correlation was not as strong as with DOC concentration.

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Table 1. Mean value of water quality parameters from June 2013 to April 2014. Turb denotes water turbidity; *N* denotes sampling numbers.

~~Item~~	pH	Salinity (PSU)	Turb (NTU)	*N*
Jun 2013	8.54	0.40	166.20 ± 108.73	15
Aug 2013	8.63	0.37	63.13 ± 31.21	13
Feb 2014	8.35	0.70	21.33 ± 15.87	17
Apr 2014	8.67	0.43	22.24 ± 16.42	22
All	8.55	0.48	62.18 ± 79.07	67

Table 2. Positions of the fluorescence maximum peaks of the four components identified by PARAFAC ~~model~~ in the present study compared with those previously identified. Secondary excitation ~~band~~ is given in brackets.

Component No	Exmax (nm)	Emmax (nm)	Description and source	~~Label 1~~	~~Label 2~~
C1	230 (300)	425	Marine humic-like (phytoplankton degradation)	M	6
C2	255 (350)	460	Terrestrial humic-like	A and C	1 and 4
C3	225 (290)	360	Autochthonous tryptophan-like	T	
C4	220 (275)	320	Autochthonous tyrosine-like	B	8

Fluoresence peaks were named as ~~Label 1~~ by Coble et al. (1996, 1998) and Zhang et al. (2010, 2011), while as ~~Lable 2~~ by Stedmon and Markager (2005).

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

Table 3. Mean values of DOC concentration and CDOM absorption coefficients groups ~~with~~ different seasons. S_R: the spectral slope ratio of $S_{275\text{–}295\,\text{nm}} : S_{350\text{–}400\,\text{nm}}$.

~~Item~~	a(254) m^{-1}	a(280) m^{-1}	a(350) m^{-1}	S_R	DOC ~~mg^{-1} L^{-1}~~	N
Jun 2013	38.39 ± 9.23	25.98 ± 6.38	5.73 ± 1.68	1.29 ± 0.16	31.84 ± 14.67	15
Aug 2013	29.71 ± 4.73	19.36 ± 2.91	5.82 ± 0.81	0.96 ± 0.22	32.83 ± 14.78	13
Feb 2014	52.88 ± 18.13	34.62 ± 11.54	6.36 ± 2.17	1.18 ± 0.11	55.04 ± 20.00	17
Apr 2014	34.43 ± 11.38	22.45 ± 7.36	4.17 ± 1.49	1.32 ± 0.13	30.86 ± 10.91	22
All	39.08 ± 14.73	25.73 ± 9.58	5.40 ± 1.84	1.21 ± 0.20	37.60 ± 18.05	67

Table 4. Correlation coefficients (R) and significance levels (p) of the linear relationships between CDOM absorption, DOC, salinity and ~~fluorescence~~ components.

	a(254)	a(280)	a(350)	DOC	Salinity	C1	C2	C3	C4
DOC	0.711**	0.646**	0.294*	1.000**					
Salinity	0.650**	0.579**	0.159	0.965**	1.000**				
C1	0.850**	0.875**	0.873**	0.496**	0.383**	1.000**			
C2	0.677**	0.686**	0.885**	0.414**	0.270*	0.796**	1.000**		
C3	0.452**	0.417**	0.134	0.648**	0.685**	0.267*	0.103	1.000**	
C4	−0.040	−0.016	0.078	−0.101	0.135	0.084	0.069	0.225	1.000**

* $p < 0.05$ level; ** $p < 0.01$ level.

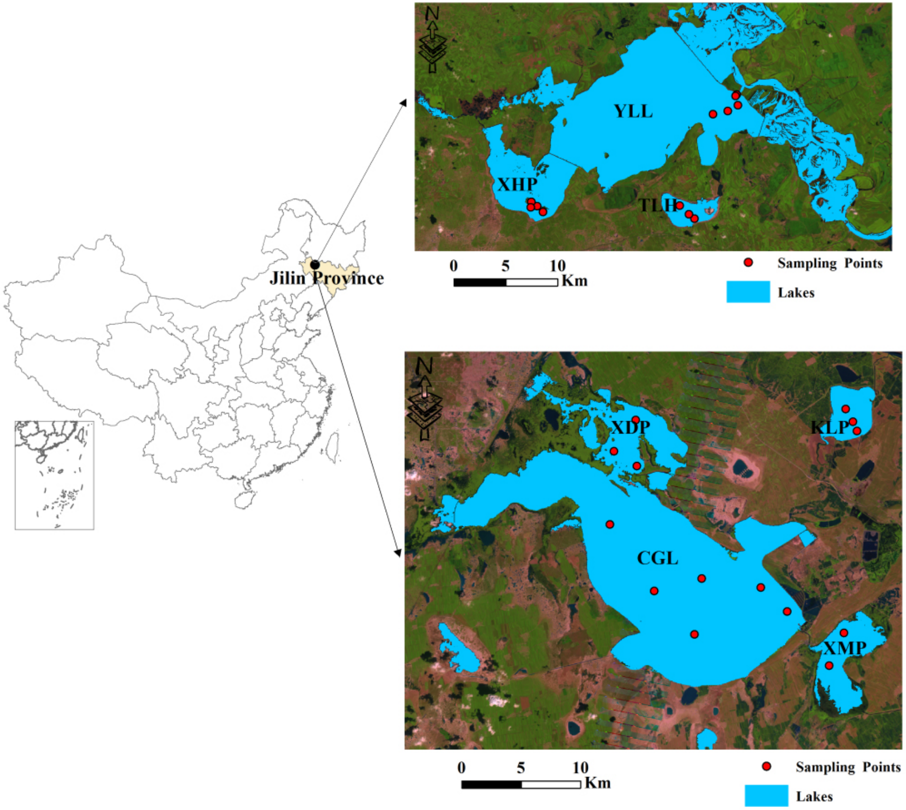


Figure 1. Locations of the water sampling sites for 7 lakes in the western part of Jilin province, Northeast China.

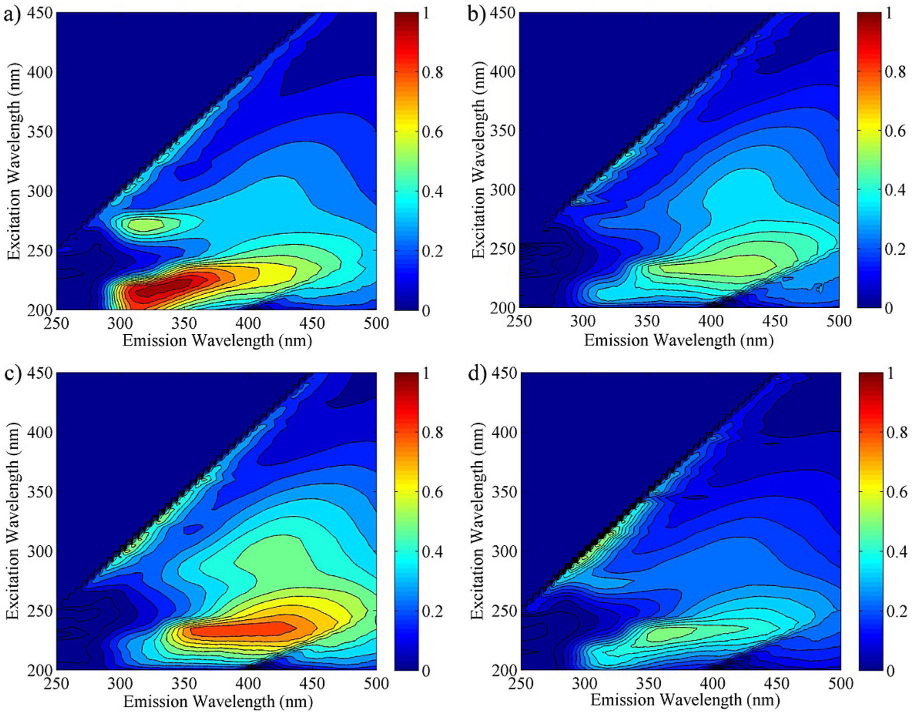


Figure 2. Examples of EEMs for one water sample from Xindianpao Lake in the western part of Jilin province~~ from~~ different seasons **(a)** June 2013; **(b)** August 2013; **(c)** February 2014; **(d)** April 2014 (~~Raman: nm^{-1}~~).

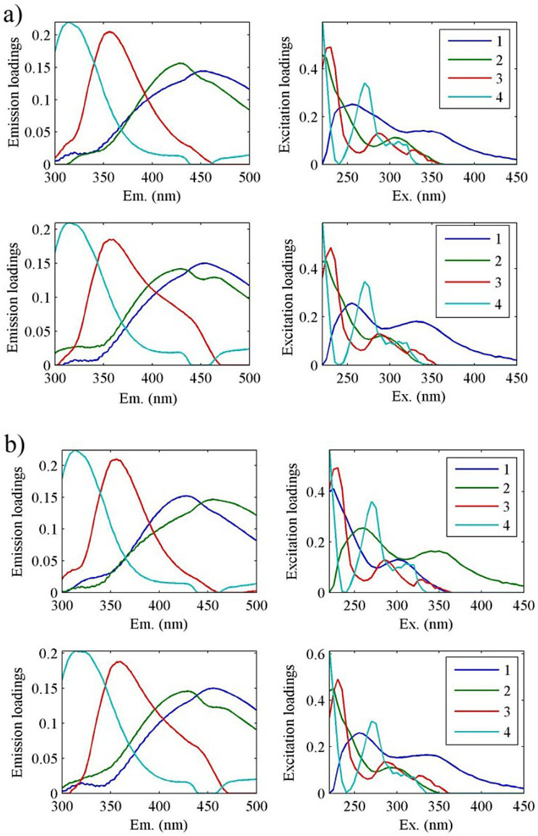


Figure 3. ~~Results from split-half analysis (1–2 left; 3–4 right) in PARAFAC models~~. The plots represent spectral shapes of the excitation and emission loadings from the two halves (1–2; 3–4 split-half analysis) modeling.

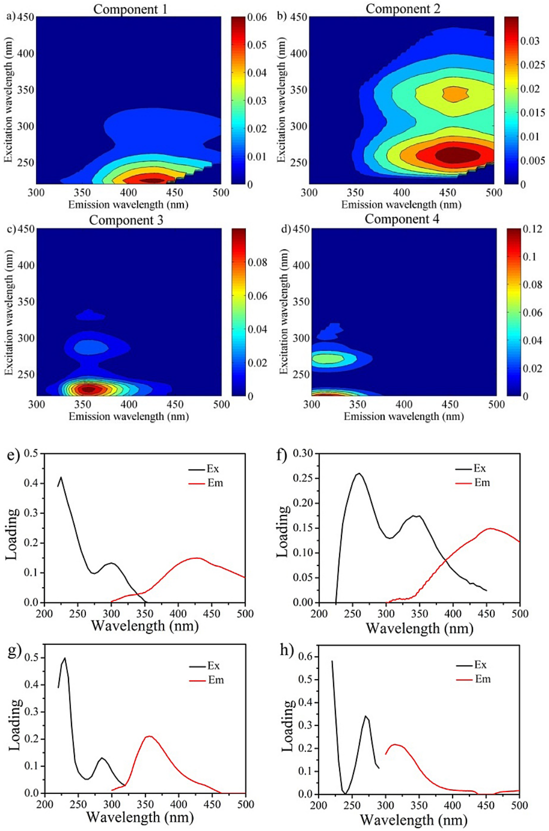
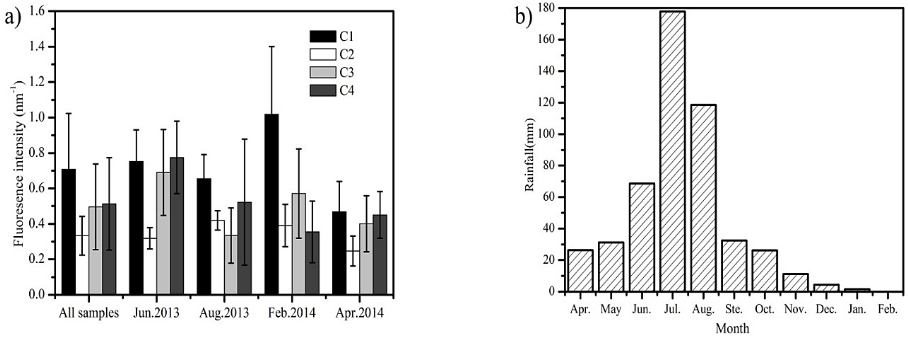


Figure 4. ~~The PARAFAC model output showing fluorescence signatures of the four fluorescence components (**a**–**d**), the contour plots represent spectral shapes of excitation and emission. The line plots represent split-half validation results (**e**–**h**): excitation (left) and emission (right) spectra~~.



~~**Figure 5. (a)** Seasonal variation of F_{max} values of the four components (C1, C2, C3 and C4) for lakes in the western part of Jilin province; **(b)** monthly variation of rainfall for the lakes in the western part of Jilin province from April 2013 to February 2014. The error bar represents SD.~~

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
I◀
▶I
◀
▶
Back
Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

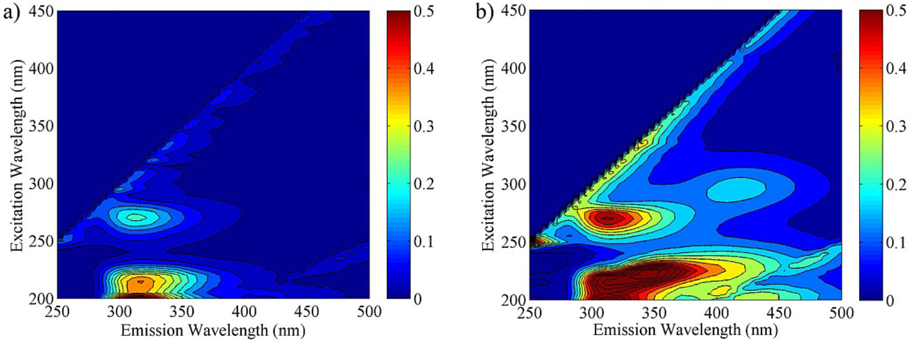


Figure 6. Representative examples of EEMs for **(a)** lake ice-melt~~ed~~ water sample, and **(b)** rainwater CDOM in the western part of Jilin province (Raman: nm^{-1}).

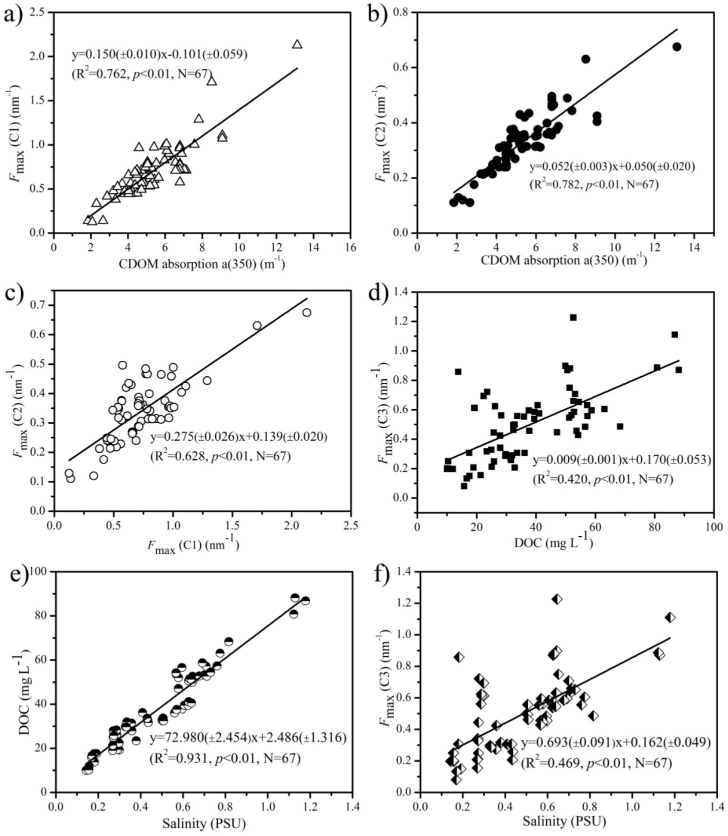


Figure 7. Relationships between CDOM absorption coefficient a(350) with **(a)** $F_{\max}$ (C1), **(b)** with $F_{\max}$ (C2), **(c)** peak $F_{\max}$ (C1) vs. $F_{\max}$ (C2), **(d)** peak $F_{\max}$ (C3) vs. DOC, **(e)** salinity vs. DOC, **(f)** salinity vs. $F_{\max}$ (C3).

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
◀▏ ▕▶
◀ ▶
Back
Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion