



New insights into the variation of dissolved organic matter components in different latitudinal lakes of northeast China

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Abstract

Dissolved organic matter (DOM) plays a key role in the carbon cycle of freshwater ecosystems. We explored DOM composition from six lakes encompassing a typical latitudinal gradient of lakes in northeast of China (41.9°N–48.6°N). Generally, the relative contents of protein-like components decreased while humic-like components increased with increasing latitude. Additionally, the results from modified two-dimensional correlation spectroscopy indicated that humic-like components with relatively simple structure were formed earlier than those with relatively complex structure. Furthermore, structural equation models suggested that at higher latitude, lake DOM tended to stabilize as protein-like component was transformed into humic-like component. We also found that nutrients (e.g., nitrogen and phosphorus) were key factors affecting DOM composition in low-latitude lakes, whereas temperature was the key factor affecting DOM composition in high-latitude lakes. In conclusion, this study identified the factors which explained the observed latitudinal pattern of DOM in northeast lakes of China. This study provided a theoretical basis for DOM management of lakes at different latitudes.

Dissolved organic matter (DOM), the primary form of organic carbon in lake ecosystems, plays a critical role in global carbon cycle, including supplying energy to support the aquatic food web and microbial metabolism (Cole et al. 2007; Battin et al. 2009; Mladenov et al. 2011). In addition, it is an essential carbon and nutrient source for heterotrophic bacterioplankton and fish in freshwater aquaculture. DOM is composed of different organic compounds with particular biogeochemical reactivities and ecological functions (e.g., proteins, humic acids, and carbohydrates of various molecular sizes). The traditional view considered that humic matter as stable DOM whereas nonhumic DOM was thought to be labile DOM. Additionally, DOM in water can lead to unpleasant odor and produce harmful disinfection byproducts during drinking water treatment processes (Zhou et al. 2016). In particular, it is difficult to remove humic acid and reduce the secondary pollution (e.g., membrane fouling) in lakes simultaneously (Liu et al. 2011). Besides, humic-like components also play an essential role in the complexation and transportation of metal ions such as Cu^{2+} , Al^{3+} , Mg^{2+} , and Cd^{2+} in freshwater systems (Yan and Korshin 2014; Yuan et al. 2015) and humic-like component is also an important source of global warming

through the mineralization of dissolved organic carbon (Zhang et al. 2011). Therefore, there is a growing interest to identify the fate of DOM components and environmental factors associated with DOM in lakes.

The composition and transformation of DOM are influenced by a variety of processes and factors, physical (e.g., climate and inflow rate), chemical (e.g., photodegradation and redox reactions), and biological (e.g., primary production and microbial decomposition) (Dalzell et al. 2009; Garcia et al. 2015; Zhou et al. 2016). Latitude is a critical factor which can affect climate, environmental conditions, and biogeochemical cycles (Liu et al. 2013; Roth et al. 2013). Taking DOM samples at a latitude gradient should allow to compare characteristics with different climatic influences and might give conclusions on future trends caused by warming of the higher latitudes (Roth et al. 2013). It is generally accepted that autochthonous DOM production and organic matter inputs from the catchment support the largest pool of organic C in lakes. Humic-like stable DOM is often closely associated with terrestrial input (Coble 2007; Zhang et al. 2011; Osburn et al. 2012), while protein-like labile DOM is mainly from microbial sources. Moreover, labile DOM also could be transformed into humic-like components during microbial metabolism (Tranvik 1993). However, whether DOM components would transform with the changes of environmental conditions induced by latitude is largely unknown. Thus, it is necessary to define more clearly whether parts of labile DOM

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Additional Supporting Information may be found in the online version of this article.

could be transformed into stable DOM in lake ecosystems with increasing latitude. Does this pathway represent a new input of humic-like substances and do environmental conditions drive this pathway?

Fluorescent dissolved organic matter (FDOM) is often used as a representation of the DOM, which can be identified via its fluorescence, thereby permitting high-resolution measurement. Fluorescence excitation-emission matrix (EEM) combining with parallel factor analysis (PARAFAC) has been used to investigate latitude-induced changes in the dynamics of components of FDOM in aquatic ecosystem (Mladenov et al. 2011; Roth et al. 2013; Walker et al. 2013). Although EEM-PARAFAC has shown to separate different fluorescence components from FDOM, helping to trace the behaviors of individual components on a given process (Osburn et al. 2012; Lee and Hur 2016), this method still could not distinguish the internal variations of two or more fluorescence peaks in one FDOM component effectively. Thus, our understanding of inner changes of FDOM components derived from PARAFAC analysis and factors associated the composition and transformation of FDOM at different latitudes is limited. Two-dimensional correlation spectroscopy (2D-COS) is another effective technique for tracing the subtle responses of a complex mixture (e.g., DOM) under the external perturbations such as metal addition, pH, time, and temperature (Lee and Hur 2016; Noda 2016; Gao et al. 2019). Traditional 2D-COS is generally based on F_{\max} values derived from PARAFAC analysis, which is a comprehensive index that was used to compare the relative content of each FDOM component under different treatments. Here, for the first time, the inner variations or intervariations of FDOM components were analyzed by the modified 2D-COS which was based on fluorescence intensity of excitation loadings derived from the PARAFAC results. This modified 2D-COS can effectively distinguish the changing order of multiple fluorescence peaks in a single component.

This study proposed a possible mechanism that the transformation of protein-like components may be an important source of humic-like components in lake ecosystems with increasing latitude. Six lakes at different latitudes in northeast of China were selected as a representative to investigate the properties of DOM under different environmental conditions. The purposes of this work were to: (1) assess the composition of DOM at different latitudes, (2) identify key environmental factors related to DOM composition at different latitudes, and (3) explore the mechanism that whether latitude affects DOM transformation directly or indirectly mediated by other environmental factors. We investigated the changes of DOM composition and transformation under latitude perturbation using EEM-PARAFAC and modified 2D-COS analysis, then tested whether latitude influenced DOM transformation directly or indirectly by structural equation models (SEMs). The study is expected to help improve our understanding of DOM in lakes with different latitudes; it may also provide the theoretical basis for management of DOM in lakes.

Materials and methods

Sampling area

A total of 152 valid samples were collected from six lakes in northeastern China in 2011. This area covered a latitude transect from 41.9°N to 48.6°N. Short, hot summers and very cold winters characterize the continental climates in this area. Detailed geographical information of sampling lakes is shown in Supporting Information Table S1. Surface-water samples were collected in 5-liter acid-cleaned bottles and stored at 4°C in the dark until laboratory measuring. Samples were transported to the laboratory immediately and filtered through a prewashed 0.22- μ m Millipore filter. DOM optical measurements were made within 3 d and all laboratory measurements were completed within 1 week. Detailed physicochemical data of sampling sites are shown in Supporting Information Table S2.

DOM characteristics

Fluorescence EEMs were measured in a 1 cm path length quartz cuvette using a Hitachi F-7000 fluorescence spectrophotometer (Hitachi High Technologies, Japan) at room temperature ($20 \pm 2^\circ\text{C}$). To weaken the inner filter effects, the concentrations of all DOM samples were diluted to 10 mg C L⁻¹ before the fluorescence scanning (Yu et al. 2010). The inner-filter effects were corrected for absorbance by the multiplication of each value in EEMs with a correction factor, based on the premise that the average path length of the absorption of excitation and emission light was one-half of the cuvette length. The correction is expressed mathematically as $F_{\text{IFE}} = F_{\text{Corr}} \times 10^{(A_{\text{Ex}} + A_{\text{Em}})/2}$, where F_{IFE} and F_{Corr} are the corrected and uncorrected fluorescence intensities, respectively, and A_{Ex} and A_{Em} are the absorbance values at the current excitation and emission wavelengths, respectively (Kothawala et al. 2013; Cui et al. 2016). Fluorescence intensities were measured at excitation wavelengths of 200–600 nm in 5 nm increments and emission wavelengths of 200–600 nm in 2 nm increments, with a scanning speed of 2400 nm min⁻¹. The excitation and emission slit widths were both set to 5 nm. Water Raman scatter were calibrated by subtracting fluorescence EEMs of Milli-Q water blanks. Rayleigh scatter effects were eliminated according to the approaches recommended in the decomposition routines for the EEM toolbox (Murphy et al. 2013). The EEMs were normalized to the daily determined water Raman-integrated area maximum fluorescence intensity ($350_{\text{Ex}}/397_{\text{Em}}$).

Multiple models analysis

EEM-PARAFAC

PARAFAC models were developed with DOMFluor toolbox (www.models.life.ku.dk) in MATLAB 2013a (Mathworks, Natick, MA) following the procedure described by Stedmon and Bro (2008). The toolbox was used not only to distinguish the possible outliers from the pixels in the EEM spectroscopy, but also to identify the agreeable number of components (Guo et al. 2018). The excitation and emission wavelengths

were restricted to 220–490 nm and 250–550 nm, respectively. Samples from six different latitudes were modeled with PARAFAC independently. This study aimed to explore whether the PARAFAC components are same in different latitudinal lakes. Thus, we decided to run a different PARAFAC analysis for each of the six lakes, rather than ran for all the lakes together. However, if we create a model based on all the lakes together, it will cover the difference in fluorescent peak position of different lakes. The size of sample used for each of the PARAFAC model is shown in Supporting Information Table S2. There were 16–42 samples used for each of the model, while there were more than 20 samples before removing outliers. Stedmon and Bro (2008) indicated that it is better to have more than 20 samples for PARAFAC. Although some models in this study have no more than 20 samples (16), our samples at different sites can reflect the overall nature of each lake. Leverage was used to examine the outliers in the model, the samples with significantly higher leverage than other samples were removed. In the PARAFAC model, 2–8 components can be computed and split-half analysis was applied to identify the number of components (Supporting Information Fig. S2). The results of split-half validation were great, suggesting that different PARAFAC analysis could effectively reflect the properties for each of the six lakes. The Tucker's congruence coefficient of all components was > 0.99 , indicating that EEM-PARAFAC model can be applied to analyze DOM changes resulting from latitude-driven for all water sample sites. The $F_{\max}\%$ values were used to compare fluorescence of a single component across different latitudes samples (Ohno et al. 2008; Mangalgi et al. 2017). Meanwhile, the concentration of all DOM samples was diluted to a same level at 10 mg C L^{-1} to make the comparison valid. Peak-picking method was used to distinguish the two peaks with the different excitation wavelength in one PARAFAC component. $I_{\text{Humic}}/I_{\text{protein}}$ was used to characterize the humification degree of DOM in this study (Zhu et al. 2018).

Modified 2D-COS

The modified 2D-COS analysis was carried out on the excitation loadings data of PARAFAC components to compare the dynamic changing orders in mini-peaks (with one maximum emission wavelength but two maximum excitation wavelengths) of PARAFAC components. Detailed information about these mini-peaks are shown in Supporting Information Table S3. This modified fluorescence 2D-COS technology, based on excitation loadings, can observe more subtle changes within each component, which could not achieve through traditional fluorescence 2D-COS based on F_{\max} . Latitude was applied as an external perturbation in the present study. Detailed information about 2D-COS analysis are described in Supporting Information Text S1.

Structural equation models

SEM is a priori approach offering the capacity to visualize the causal relationships between variables by fitting data to the models representing causal hypotheses (Wu et al. 2017; Zhu

et al. 2019). SEMs were used to evaluate the direct or indirect effects of latitudes on DOM composition and transformation between different fluorescent components. We used nonsignificant chi-square test ($p > 0.05$), high goodness-of-fit index (GFI > 0.90), low Akaike information criteria (AIC < 0.05), and low root mean square errors of approximation (RMSEA < 0.05) to show the overall goodness of fit for SEMs.

Physicochemical analyses

Water temperature (T), dissolved oxygen (DO), pH, and chlorophyll a (Chl a) were directly measured by the multi-parameter water quality analyzer (Hydrolab DS5, Hach Company, U.S.A.). Total nitrogen (TN), ammonia nitrogen ($\text{NH}_4^+\text{-N}$), nitrate nitrogen ($\text{NO}_3^-\text{-N}$), nitrite nitrogen ($\text{NO}_2^-\text{-N}$), total phosphorus (TP), dissolved phosphorus (DP), 5-d biochemical oxygen demand (BOD_5), and KMnO_4 index (KI) were analyzed according to the standard methods (State Environment Administration of China 2002). In the present study, the above environmental parameters were only used to analyze the relationship between them and DOM components. In addition, there were no obvious differences in land uses and human activities between sampling lakes, thus these factors were not considered for influencing DOM transformation.

Statistical analysis

Physicochemical data were analyzed with one-way ANOVA and t -test by SPSS 22.0 and ORIGIN 2016. 2D-COS analysis was performed using 2D Shige software (Kwansei-Gakuin University, Japan). Canoco 5.0 was used for determination of the relationship between environmental factors and DOM components. Redundancy analysis (RDA) was recommended when a detrended correspondence analysis suggested that DOM composition exhibit a linear response to the environmental factors (Šmilauer and Lepš 2003). Monte Carlo permutation model was used to test the statistical significances of axis and individual parameters through 499 unrestricted permutations (Kim et al. 2016). SEMs were performed by AMOS 20.0 software (IBM Corporation Software Group, Somers, NY) using the maximum-likelihood estimation method.

Results and discussion

Variations in DOM components under latitude perturbation

To assess the latitudinal pattern of DOM composition, all the EEM data from six latitudes were modeled by PARAFAC, respectively. Three fluorescence components (C1, C2, and C3) were identified in low-latitudinal lakes (41.9°N – 43.9°N) by EEM-PARAFAC analysis (Supporting Information Fig. S1 and Table S3). In addition, another component (C4) was identified in high-latitudinal lakes (45.8°N – 48.6°N). We used split-half analysis to confirm the appropriate number of DOM components (Stedmon and Bro 2008). Based on the results of split-half analysis, three components models were considered to

satisfy the validation at low latitudes, while four components models satisfied the validation at high latitudes (Supporting Information Fig. S2). Component C1 was characterized as representing protein-like substance, which may be associated with a biological production and activity of microorganisms (Fig. 1e) (Stedmon et al. 2003; Hudson et al. 2008; Xiao et al. 2019). Component C2 was similar to a fulvic acid-like substance (Fig. 1f) (Yamashita and Jaffé 2011; Yuan et al. 2017). Component C3 corresponds well to the terrestrial humic-like fluorophores (Fig. 1g) (Stedmon and Markager 2005; Murphy et al. 2011; Tanaka et al. 2014). Component C4 was characterized as representing the UVC humic-like substance, which had a simpler structure than component C3 (Fig. 1h) (Ishii and Boyer 2012). The relative contribution of protein-like component C1 decreased with latitude (Fig. 1a). Meanwhile, the relative contribution of humic-like components (C2–C4) increased from 55% to 81%, suggesting that the humification degree of DOM increased with latitude.

There were two or more peaks in each DOM component, we called those mini-peaks, which were neglected in previous studies. In the present study, we described the influence of latitude on the intensities of these mini-peaks in detail (Supporting Information Table S4). It is observed that the intensities of peak A2 and B2 were negatively correlated with latitude ($r^2 = 0.60$, 0.62 ; $p < 0.05$) (Fig. 1b,c). In contrast, the intensity of peak E1 was positively correlated with latitude ($r^2 = 0.74$, $p < 0.05$)

(Fig. 1d). However, there were no significant correlations between latitude and intensities of other peaks. In the present study, the modified 2D-COS based on excitation loadings effectively distinguished these peaks with two excitation maxima but one emission maximum in one component (e.g., peak A1 and A2 in component C1) under latitude perturbation. 2D-COS and hetero 2D-COS were used to identify the order of changes in these mini-peaks which significantly changed with latitude (i.e., A₂, B₂, E₁, D₁, and D₂). Detailed rules about 2D-COS analysis are shown in Supporting Information Text S1.

Figure 2a,b shows the synchronous and asynchronous maps of the hetero-2DCOS analyzed by excitation loadings of component C1 and C2. In synchronous hetero-2DCOS, peak A₂ was positively correlated to the peak B₂. However, in the asynchronous map (Fig. 2b), there were no significantly positive or negative signs between peak A₂ and B₂. Thus, the order of changes between peak A₂ and B₂ cannot be determined. Figure 2c,d shows the synchronous and asynchronous maps of the hetero-2DCOS analyzed by excitation loadings of component C3 and C4. In synchronous hetero-2DCOS, peak E₁ was negatively correlated to peak D₁, while positively correlated to peak D₂. Inversely, in asynchronous map, peak E₁ was positively correlated to peak D₁, while negatively correlated to peak D₂. Thus, the variation of the peaks with the increase of latitude follows the order peaks D₁ and D₂→E₁. In order to identify the order of changes between peak D₁ and D₂,

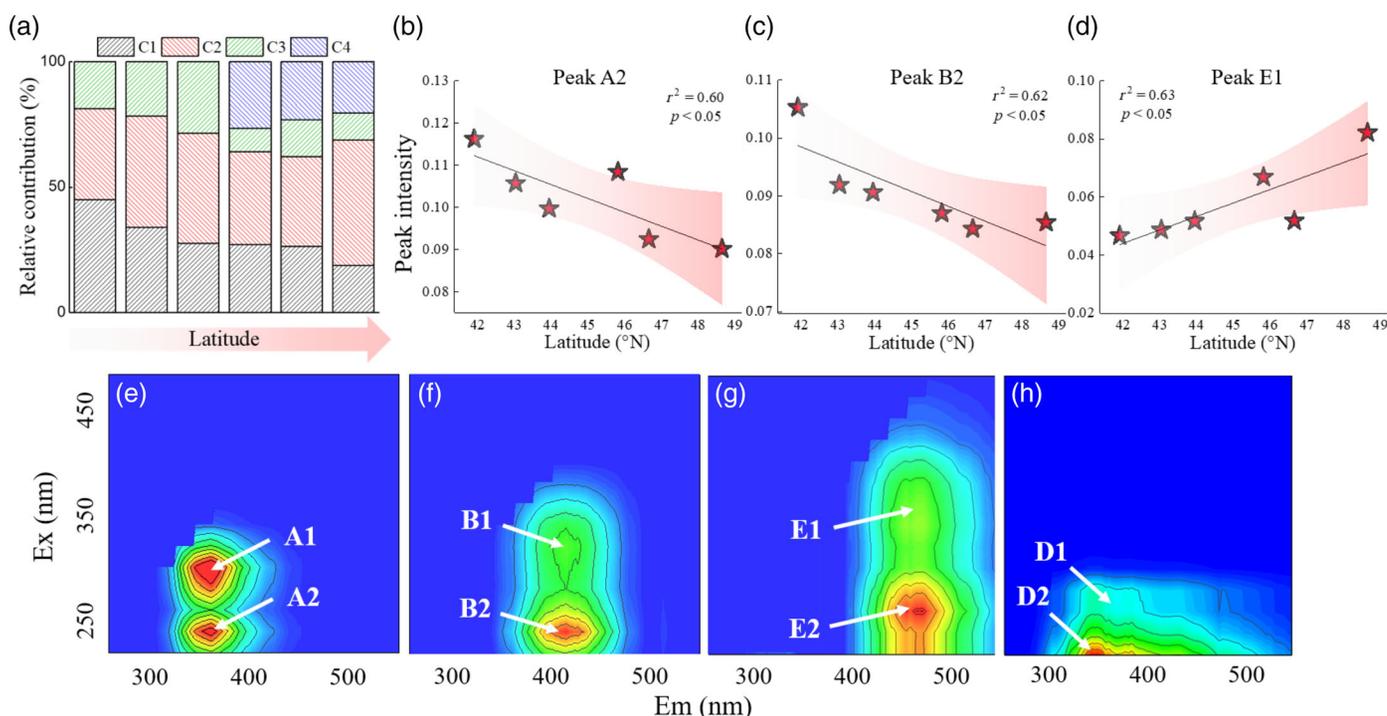


Fig. 1. Variations of DOM components in different latitudinal lakes. **(a)** The relative contribution of DOM components in different latitudes. C1–C4 represents DOM components 1–4. **(b–d)** Variations in mini-peaks of DOM components with the increase of latitude. The relationships between all mini-peaks and latitudes were tested. Only three of them (peak A₂, B₂, and E₁) changed significantly with latitude. **(e–h)** DOM components C1–C4 derived from PARAFAC. [Color figure can be viewed at wileyonlinelibrary.com]

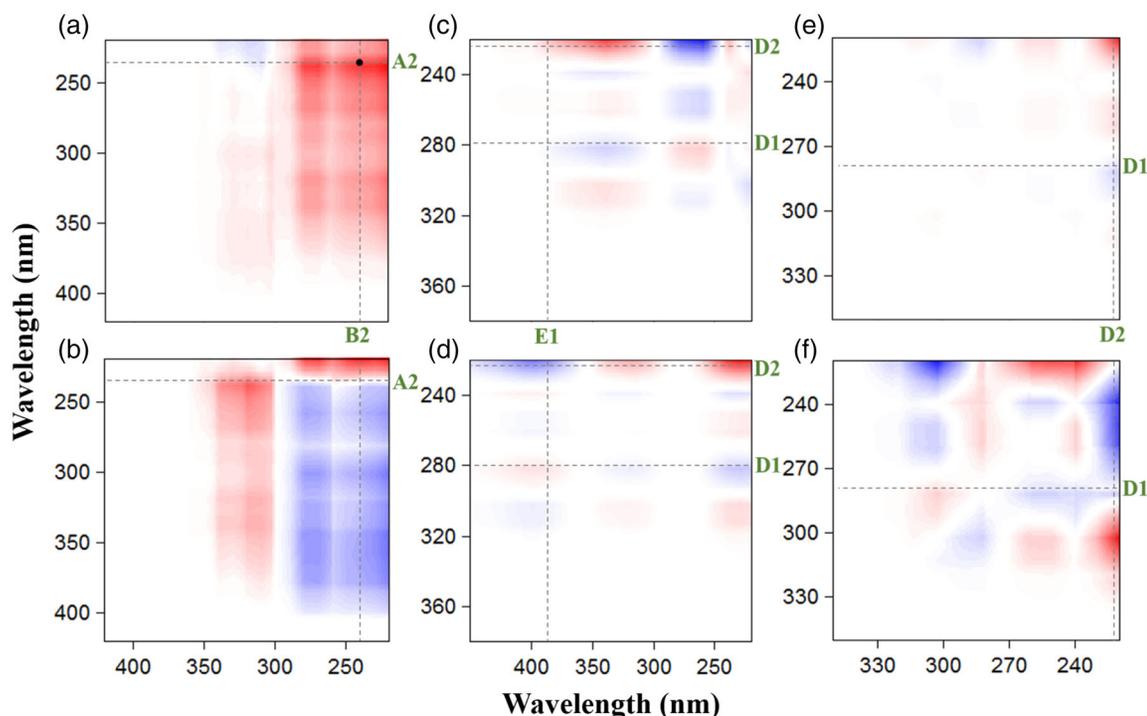


Fig. 2. Synchronous and asynchronous maps of 2D-COS and hetero 2D-COS of PARAFAC components under latitude perturbation. Red represents positive correlations and blue represents negative correlations. (a) Synchronous hetero-map, C1 vs. C2. (b) Asynchronous hetero-map, C1 vs. C2. (c) Synchronous hetero-map, C1 vs. C2. (d) Asynchronous hetero-map, C1 vs. C2. (e) Synchronous map of C4. (f) Asynchronous map of C4. [Color figure can be viewed at wileyonlinelibrary.com]

the synchronous and asynchronous spectra of component C4 are described in Fig. 2e,f. Peak D1 was negatively correlated to peak D2 both in synchronous and asynchronous spectrum. The peaks in component C4 follow the order peak D2→peak D1 with the increase of latitude. Based on the above results, the substances are formed as latitude increased in the following order: peak D2→D1→E1, while the order of decomposed substances (peak A2 and B2) cannot be determined. These results proved that humic-like components with relatively labile structure (peaks D1 and D2) were appeared earlier than those with relatively stable structure (peak E1) with the increase of latitude.

A possible transformation mechanism for DOM components

It is generally accepted that terrestrial input is one of the main sources of relatively complex DOM (Zhou et al. 2016). Furthermore, DOM can be utilized and transformed by many in-lake processes, including photodegradation, microbial respiration, and flocculation (Wachenfeldt et al. 2008; Cory et al. 2007; Evans et al. 2017). Generally, microbial communities only produce part of DOM with low-molecular weight and aromatic structure (e.g., amino acid or protein-like components). Moreover, the previous study demonstrated that humic matter might be formed in the aquatic environments from labile organic carbon compounds such as glucose (Tranvik 1993). Microorganisms could also produce complex

DOM and in fact could aggregate them (Ogawa et al. 2001). Additionally, labile DOM can also be transformed into stable DOM by flocculation. Based on previous studies, we proposed a hypothesized mechanism that the transformation from labile DOM to stable DOM may be an important source of humic-like substances, especially in high-latitude lakes. In different latitudinal lakes, the distribution of DOM components may be regulated by nutrient and environmental conditions to meet the needs of microbial growth and metabolism. In the next section, we tested our hypothesis about the transformation mechanism of DOM in different latitudes and typical environmental factors related with DOM transformation.

We constructed SEMs to test the causal relationships among the eight peaks in four DOM components (Fig. 3). As for the samples from different latitudes (Fig. 3a,c), some similar results were found that peaks A2, B2, and E2 had significantly positive influences on peaks A1, B1, and E1, respectively. Meanwhile, peak B2 had a strong positive effect on peak E2 at both low and high latitudes. These results revealed that at least part of the increase in stable DOM (e.g., humic-like component) derived from increasing latitude may be associated with the degradation of labile DOM (e.g., protein-like component). In addition, the causal relationships became more complex in high latitudes than those in low latitudes (Fig. 3c). In high latitudes, peak B2 directly affected peak E1, meanwhile peak B2 could also indirectly

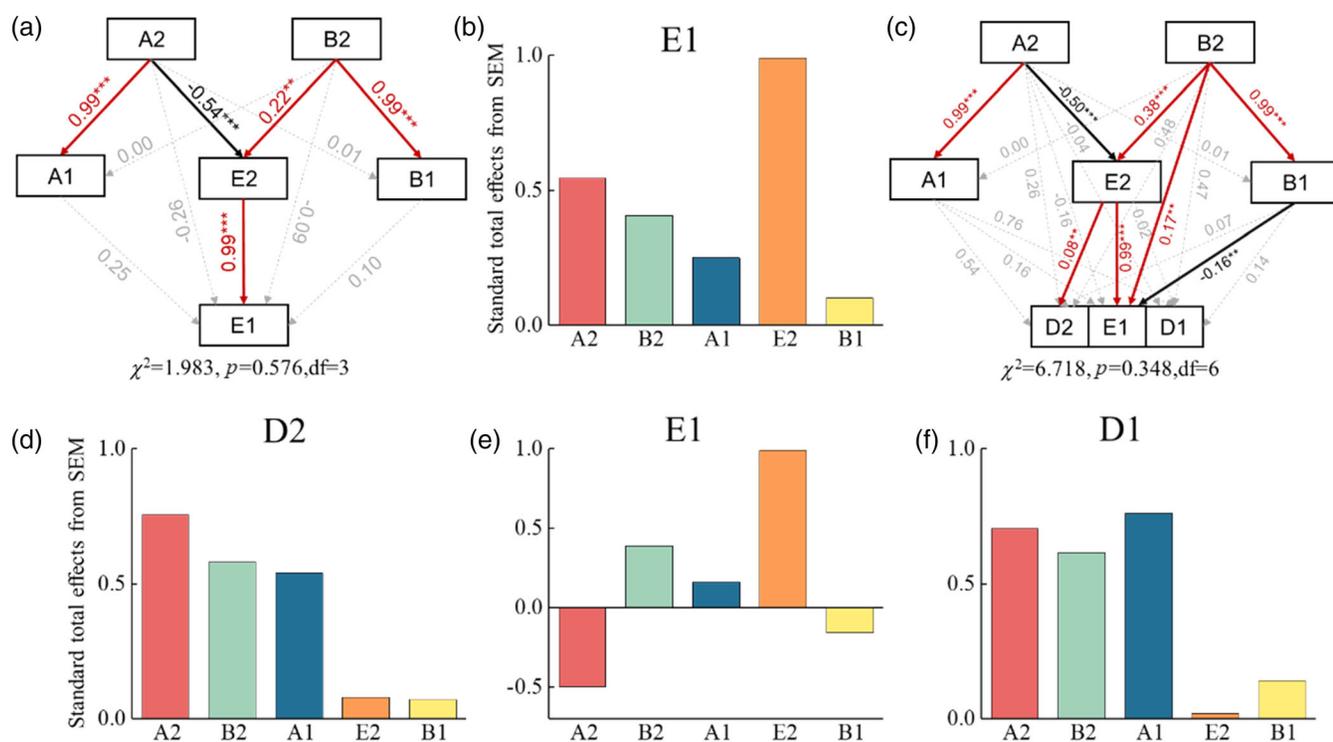


Fig. 3. Transformation from labile DOM to stable DOM verified by SEMs. (a) SEM of DOM samples in low latitudes. (b) Standard total effects (direct plus indirect effects) on peak E1 derived from SEM in low latitude. (c) SEM of DOM samples in high latitudes. (d–f) Standard total effects (direct plus indirect effects) on peaks D2, E1, and D1 derived from SEM in high latitude. (a, c) Red and black arrows indicate positive and negative relationships, respectively. Solid line and short dash line indicate significant ($p < 0.05$) and nonsignificant relationships ($p > 0.05$), respectively. Numbers adjacent to arrows are standardized path coefficients, analogous to relative regression weights, and indicative of the effect size of the relationship. Paths with coefficients nonsignificant different from 0 ($p > 0.05$) are presented on gray. * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$. Goodness-of-fit statistics for each model are shown below the figure. [Color figure can be viewed at wileyonlinelibrary.com]

affect peak E1 through influencing peak E2 and B1. In addition, peaks A2 and B2 also had significantly indirect influences on peaks D2 and E1 through influencing peaks B1 and E2, suggesting that peaks B1 and E2 may be critical intermediate products during the evolution along the gradient of latitudes. As indicated by the standardized total effects from SEMs, E2 is the most important predictor of E1 in both low and high latitudes (Fig. 3b,e). In addition, A2, B2, and A1 were three main sources of the formation of new component C4 (D1 and D2) (Fig. 3d,f), which proved the conversion of labile DOM into stable DOM with the increase of latitude. Peaks A2 and E2 are two main sources of peak E1 in both low and high latitudes (Fig. 3b). However, peak A2 became a negative factor influencing peak E1 in high latitudes in comparison with the causal relationships in low latitudes. Therefore, the significant decrease in peak A2 with the increase of latitude is a major cause of elevated peak E1. In addition, we also confirmed that protein-like (peak A1 and A2) and microbial humic-like (peak B2) substances are the main sources of the new component C4 at high latitudes. Because, protein-like and microbial humic-like components are easily removed by microbial degradation processes (Cohen et al. 2014; Yu et al. 2015). Overall, these results are consistent with our conjecture that labile

DOM could be transformed into stable DOM with the increase of latitude.

Although SEMs can partly explain the transformation in DOM components, there may be also many other factors influencing the fate of DOM in lake processes. For example, increasing the amount of carboxyl and hydroxyl groups on certain DOM type could lead to shift fluorescence to longer wavelengths. Moreover, biological activity during lake transport or in the coastal ocean can result in production of new fluorescent materials, including a newer humic-like material. Previous reports also suggested that photochemical degradation processes cause a gradual shift in peak C position toward shorter wavelengths (blue-shift) with a concomitant decrease in fluorescence intensity (Coble et al. 2014).

Environmental factors affecting DOM transformation

The increasing tendency of humification degree of DOM (from 2.97 to 5.47) with the increase of latitudes promoted us to identify the environmental factors affecting DOM transformation. RDA was performed to investigate the relationships between environmental factors and DOM components. All of the canonical axes were significant ($p < 0.05$), demonstrating that these environmental factors may be important in explaining the

variation of DOM components. The variation of the DOM-environmental factors relation explained by the first two canonical axes was 55.4% and 45.2% for low and high latitudes, respectively. RDA results demonstrated that DOM components are significantly ($p < 0.05$) correlated with nutrient concentrations (e.g., NO_3^- , TP, DP, and BOD_5) in low latitudes (Fig. 4a), while environmental conditions (e.g., temperature, KI, and Chl *a*) become more important in influencing DOM composition in high latitudes (Fig. 4b). However, from the RDA results, no direct significant correlations were observed between latitude and DOM components. If indeed the relationships between latitude and DOM components in this work are reflective of an indirect causal connection, further study about critical nutrient conditions and environmental factors in these lakes will enhance our understanding of DOM composition and transformation.

SEMs were used to further confirm the causal relationships among latitudes, DOM components, and other environmental factors deeply (Fig. 5). Based on the results from RDA (Fig. 4), environmental factors, which were most closely related to each DOM component, were selected for the SEM analysis (e.g., TN and NO_3^- for C1, BOD_5 and temperature for C2 at low latitudes, etc.). The hypotheses in SEMs were based on the principle that the different environmental conditions at different latitudes may directly influence DOM components.

Nonsignificant chi-square test ($p > 0.05$), high GFI (> 0.90), and low RMSEA (< 0.05) all showed the overall goodness of fit for SEMs. As for low-latitude lakes, latitude had significantly indirectly negative effects on component C1 mediated by TN

then NO_3^- (Fig. 5a). This finding was consistent with the previous report that nutrient can drive microbial transformation or production of DOM (Williams et al. 2010). In contrast to low latitude, DO became a mediator of latitude affecting component C1 at high latitudes (Fig. 5b). It may be due to that microbes need oxygen to make protein-like component convert into inorganic nitrogen to feed their own nutritional needs. BOD_5 was the main factor affecting component C2 in low latitudes, while latitude had significantly direct effects on component C2 in high latitudes, without mediated by environmental factors (Fig. 5c,d). Previous studies have demonstrated that C2 is similar to a microbial oxidized component (Cory and Mcknight 2005) and a phytoplankton degradation release humic-like component due to microbial activities (Zhang et al. 2009). Therefore, this phenomenon can be explained that microbial metabolism is more vigorous in low latitudes than that in high latitudes. As for low latitudes, BOD_5 , TP, and TDP are mediators of latitude effects on component C3 (Fig. 5e), which may be that humic matters associated with TDP, as previous studies reported (Jones et al. 1988). However, in comparison with low latitudes, temperature and Chl *a* became two important factors affecting the formation of humic-like substance in high latitudes (Fig. 5f,g). Interestingly, Chl *a* had a possible effect on component C4 (Fig. 5g), while a negative effect on C3 (Fig. 5f). It is known that photosynthesis represents the largest source of labile DOM to the lake (Urban et al. 2005). This phenomenon reflected that photosynthesis contributed to the formation of C4, but not to C3. Furthermore, temperature was the most highly influential factor

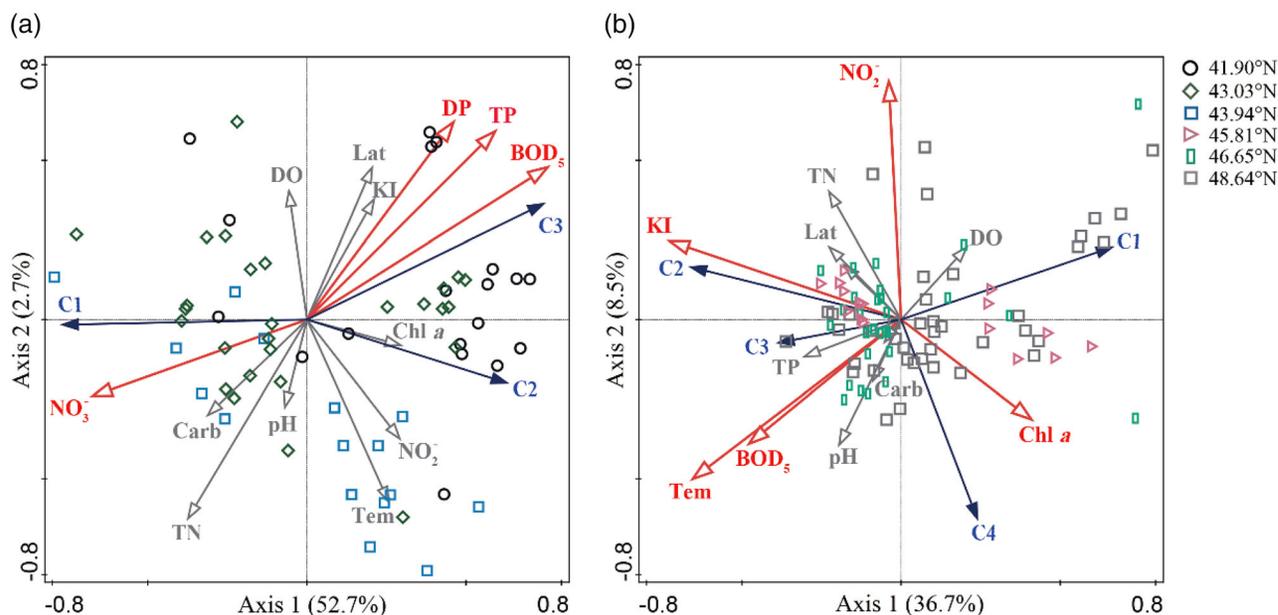


Fig. 4. Ordination plot of RDA with environmental factors and DOM components. The position of the sampling sites reveals their distribution over latitude ranges. **(a)** Low latitudes (41.90°N–43.94°N) and **(b)** high latitudes (45.81°N–48.64°N). Red arrows represent significant correlation with DOM components ($p < 0.05$), gray arrows represent nonsignificant correlation with DOM components ($p > 0.05$). Carb, carbonate; Lat, latitude; Tem, temperature. [Color figure can be viewed at wileyonlinelibrary.com]

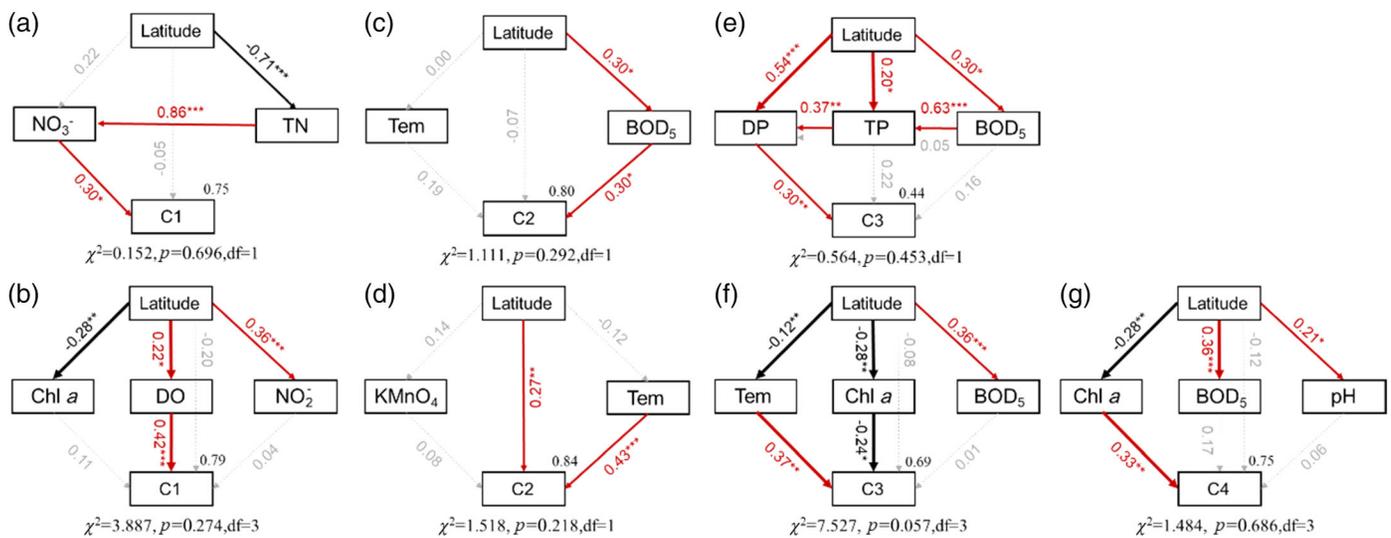


Fig. 5. Effects of latitude on DOM components. (a, c, e) DOM samples in low latitudes (41.90°N–43.94°N). (b, d, f, g) DOM samples in high latitudes (45.81°N–48.64°N). Red and black arrows indicate positive and negative relationships, respectively. Solid line and short dash line indicate significant ($p < 0.05$) and nonsignificant relationships ($p > 0.05$), respectively. Numbers adjacent to arrows are standardized path coefficients, analogous to relative regression weights, and indicative of the effect size of the relationship. Paths with coefficients nonsignificant different from 0 ($p > 0.05$) are presented on gray. * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$. The number on the top right of the component represents the percentage of component changes that can be explained by SEMs. Goodness-of-fit statistics for each model are shown below the figure. [Color figure can be viewed at wileyonlinelibrary.com]

promoting the formation of the humic components (C2 and C3) at high latitudes (Fig. 5d,f). Previous studies also found that humic-like components are abundant in cold, northern lakes (Kellerman et al. 2015). Microbial growth and metabolism process could be slowed down due to the lower temperature at high latitudes, especially in winter. Consequently, the accumulation of stable DOM is greater than the consumption of labile DOM in high latitudinal lakes. The above results from SEMs confirmed that latitude influenced DOM composition indirectly through changing complex environmental factors (e.g., temperature, nitrogen, and phosphorus).

Implications for DOM cycling in lake ecosystems

Our study suggested that latitude had significantly ($p < 0.05$) indirect effects on DOM cycling in lake systems, mediated by key environmental factors. The different relationships demonstrated in Fig. 4a,b indicated differences in the distribution of DOM components between low and high latitudes. Labile DOM in low latitude can be an important carbon source to microbial communities. DOM originating from high latitude is typically considered more humified than those from low latitude. We found that humification degree of DOM in lake ecosystem increased with latitudes and more labile fractions were transformed into humic-like matter due to the weak metabolism of microorganism and phytoplankton. Based on the relationships between environmental factors and DOM components derived from RDA and SEMs, we concluded that latitude has significant indirect effects on DOM components, mediating by influencing the concentration of nutrients (nitrogen, phosphorus, and BOD₅) in low latitudes, while

environmental conditions (temperature, Chl *a*, and DO) become important mediators in high latitudes. Consequently, these results should be interpreted that latitude is a key factor influencing DOM cycling in lake ecosystems.

Temperature plays a fundamental role in regulating biological process rates. In low latitudinal lakes (Fig. 6), temperature is relatively high, thereby metabolism is active. Thus, lots of

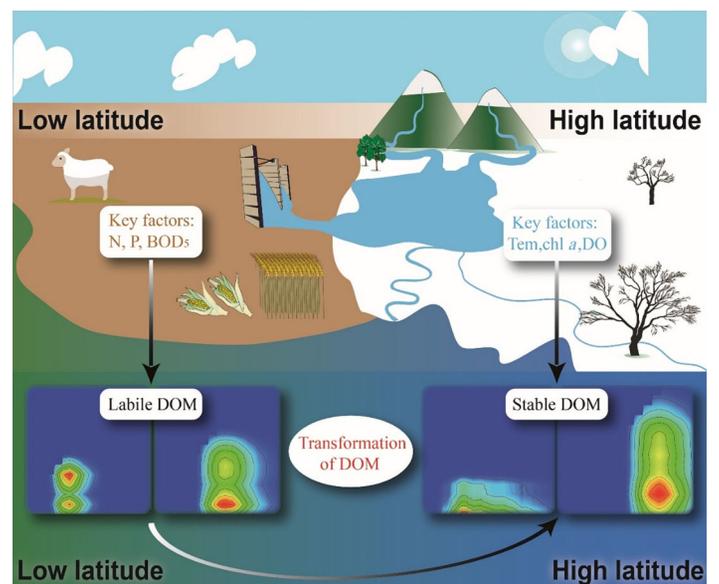


Fig. 6. Summary of the transformation between DOM components at different latitudes. Tem, temperature. [Color figure can be viewed at wileyonlinelibrary.com]

labile DOM (e.g., protein-like and microbial humic-like components) would be produced by microorganisms at low latitudes. Additionally, the influence of nutrients is also inherently linked to the accumulation of labile DOM. Kellerman et al. (2015) found that nutrient import from the catchment promotes in-lake production and transformation of DOM. However, in high latitudinal lakes, microbial growth, metabolism, and phytoplankton respiration were slowed down due to low temperature. Thus, microorganisms prefer to store DOM rather than break it down. Roth et al. (2013) also detected a higher abundance of aromatic unsaturated C=C functionalities, which might be explained by the higher abundance of refractory compounds generated due to temperature-limited decomposition.

Microorganism and phytoplankton must decompose proteins and labile humic matters to survive at low temperature. Then, the products of decomposition further form stable humic acid. From an ecological perspective, a higher level of humic substances in lakes could stimulate greater food web subsidies from energy mobilized by bacterioplankton (Jansson et al. 2007). Therefore, the latitudinal pattern of DOM transformation must be considered to predict the fate of lake DOM.

Previous studies indicated that it is difficult to accurately distinguish between the relative importance of in-lake processes (e.g., primary production, photodegradation, decomposition) and hydrological dilution, which may have collectively contributed to the observed transformation in DOM composition (Kothawala et al. 2014). In this study, we proposed that the difference in environmental conditions at different latitudes is likely to be an important driver for the transformation of labile DOM to stable DOM in complex in-lake processes. This study provides a theoretical basis for studying the changes of DOM composition with latitudes in other low temperature regions around the world. However, due to the complex conditions in lakes, it is difficult to ascertain which of the factors (environmental, geographical, or human parameters) had a greater effect on the chemical composition of DOM only by statistical analyses. In future study, the larger scale in situ experiments are needed to confirm the transformation of DOM components under different latitudes and get an increased understanding of the mechanism of DOM transformation in lakes by investigating corresponding functional groups and genes.

Conclusions

This study highlighted the latitudinal pattern of DOM composition. SEMs confirmed that at higher latitude, lake DOM tended to stabilize as protein-like component was transformed into humic-like component. Furthermore, the effect of latitude on DOM composition was indirect which was mediated by key environmental factors. In low latitudes, the higher temperature and more nutrient conditions could lead to the increase in protein-like components. In contrast, the lower temperature could lead to accumulation of humic-like

components in high latitudes. In conclusion, this study indicated that the transformation from labile DOM to stable DOM with increasing latitude may be a neglected source of humic-like substances, especially in high-latitude lakes.

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Conflict of Interest

None declared.

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